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Progress Report on the Investigation of the
Requirements for Initiation and Sustained Propagation of
Fuel-Air Explosives for the Period
 May 1, 1982 to April 30, 1983

Submitted by:

Dr. R. Knystautas
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McGill University,
 Montreal, Canada

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equivalent chemical times derived from it) provide a much more useful parameter as input to the required theories and empirical laws for the prediction of the desired requirements for initiation and propagation than the currently used induction time itself. It is the first and perhaps the most basic in the hierarchy of so-called "dynamic" detonation parameters which are parameters that depend on the rates of chemical reactions (hence "dynamic") as opposed to other detonation parameters such as the Chapman-Jouguet velocity, pressure, etc. which are derived from one-dimensional steady state equilibrium theory (hence static). Experience shows that the dynamic parameters reflect more intimately the detonation properties which are of great practical interest (e.g. initiation, transmission, detonability, etc.). In terms of critical tube diameter, the universality of the important empirical law of $d_c = 13\lambda$ has been conclusively verified in the current work. Moreover, the study of the critical tube diameter which is essentially the problem of the transmission of detonation wave from a confined circular tube to the unconfined three-dimensional geometry has been extended to other transmission geometries and the results correlated successfully again in terms of a simple empirical model. The validity of the surface energy model to correlate the detonation cell size and the critical initiation energy has been further confirmed for a broad range of explosive mixtures. This simple phenomenological model has been used to successfully link, on a quantitative basis, the detonation cell size λ with the critical initiation energy not only for hydrogen-air mixtures but for most of the common gaseous hydrocarbon fuels ranging from ethylene to the alkanes. Finally, detonability limits have also been correlated in terms of the cell size λ . Apart from the direct importance of being able to deduce detonability limits again from a relatively straight-forward procedure of measuring the cell size, there arises the further important conclusion that one cannot define detonability limits in terms of thermochemical properties of the mixture alone but that one must also define the confining geometrical boundary conditions to unambiguously define the true detonability limits for a given mixture in a configuration.

In the current program year, emphasis was also placed on the study of the problem of intense turbulent mixing between the burnt and the unburnt explosive mixture to explore specifically the environment under which rapid amplification to detonation can be achieved. The turbulent mixing was induced by having a weakly initiated flame propagate into an obstacle environment along a linear combustion tube and thereby create the intense turbulent flow field ahead of itself by virtue of fluid displacement as a result of the increase of the specific volume in an exothermic reaction. The conditions of turbulent mixing between the burnt and unburnt FAE mixture, the resulting rapid flame acceleration and the minimum necessary conditions for transition to detonation have been studied in the present program.

1. Introduction

The underlying task in FAE research as it pertains to AFOSR needs can be summarized in the following way: For a given fuel-air mixture at prescribed initial and boundary conditions, the problem is to determine the essential requirements for direct initiation and sustained propagation of a detonation wave within the FAE cloud. This implies establishing a priori on a quantitative basis the minimum required spatial extent of the activation region for initiation, the characteristic chemical time scale and the energetics required for the subsequent evolution of the detonation wave in the cloud. Our approach in the AFOSR sponsored program at McGill has been to look at the fundamental physical and thermo-chemical phenomena underlying these events from a sufficiently broad range of perspectives such that the important controlling mechanisms can be identified and thereby the events can be anticipated and controlled on a quantitative parametric basis. This approach has entailed a broad range of in-house exploratory laboratory scale experiments followed up by theoretical modelling on a phenomenological basis to lay the groundwork and establish guidelines for large scale field experiments done in collaboration with government and quasi-government research agencies which have large scale field test facilities. The present progress report describes the results that have evolved from such an approach where the activity pertinent to AFOSR needs has gone beyond the immediate in-house research work sponsored at McGill and benefited in scope, scale, level of activity and productivity by having access to and interaction

with external agencies in the manner described. Specifically, in the current program year, the results of which are being reported herein, large scale tests have been conducted in collaboration with Sandia National Laboratories, Albuquerque, New Mexico and the Defense Research Establishment Suffield, Alberta and the Norwegian Defence Construction Services, Oslo, Norway. The results of these tests form an integral part of the progress pertinent to the AFOSR program and are included herein as discrete, self-contained but intimately related sections of the progress report. The whole report is composed of individual Appendices describing the progress achieved in the various aspects of the research. All of the Appendices form part of a series of publications which have appeared recently in archive journals in the field or are being presented at international conferences as indicated on the introductory page of each study. In the next several paragraphs a summary description of the progress report is presented to assist in placing the progress reported in each Appendix in the context of the AFOSR program.

2. Measurement of Detonation Cell Size (Chemical Length Scale)
and Correlation with the Dynamic Detonation Parameters

Over the span of the past several years, extensive studies largely carried out in our laboratory have shown that the detonation cell size λ is of great fundamental significance to describe an overall chemical time scale that truly characterizes

the chemical reactions in the transient three-dimensional reaction zone of the complex multi-headed (cellular) structure of a real detonation wave itself. It is concluded that the detonation cell size λ (or the equivalent chemical times derived from it) provide a much more useful parameter as input to the required theories and empirical laws for the prediction of the desired requirements for initiation and propagation than the currently used induction time τ itself. It is the first and perhaps the most basic in the hierarchy of so-called "dynamic" detonation parameters which are parameters that depend on the rates of chemical reactions (hence "dynamic") as opposed to other detonation parameters such as the Chapman-Jouguet velocity, pressure, etc. which are derived from one-dimensional steady state equilibrium theory (hence static). Experience shows that the dynamic parameters reflect more intimately the detonation properties which are of great practical interest (eg. initiation, transmission, detonability, etc.). In last year's progress report, a limited range of cell size measurements were reported and correlated with the so-called critical tube diameter and initiation energy largely in hydrogen-air mixtures. The critical tube diameter refers to the minimum tube diameter for which a planar detonation in a particular explosive mixture will transmit without failure from a confining tube into an unconfined environment. It has been shown to be a very important dynamic detonation parameter because it can potentially be used in a number of important practical estimates in the context of the FAE problem. In the first instance, it can be used to infer

the minimum size of a detonable cloud and hence the detonability limits for a given set of boundary conditions. In the context of initiation via chemical sensitizers (FAE III concept) it can serve as a quantitative measure of the minimum spatial length scale of the activation region required for detonation initiation. Finally, the critical tube diameter has also been proposed as an alternate, more easily measurable parameter to assess shock sensitivity of a given explosive mixture instead of initiation energy which from the practical point of view is prohibitively difficult to measure for other than relatively sensitive mixtures. Nevertheless, initiation energy in itself still remains an important and basic dynamic detonation parameter; its limitation rests with the serious difficulty of its quantitative experimental measurement.

The current effort that is being reported extends the preliminary success over a limited range of conditions described in the last progress report of quantitatively correlating the cell size λ with the critical tube diameter, the initiation energy and the detonability limits. In terms of the critical tube diameter, the universality of the important empirical law of $d_c = 13\lambda$ has been conclusively verified in the current work. Moreover, the study of the critical tube diameter which is essentially the problem of the transmission of a detonation wave from a confined circular tube to the unconfined three-dimensional geometry has been extended to other transmission geometries and the results correlated successfully again in terms of a simple empirical model. The validity of the surface energy model to

correlate the detonation cell size and the critical initiation energy has been further confirmed for a broad range of explosive mixtures. This simple phenomenological model has been used to successfully link, on a quantitative basis, the detonation cell size λ with the critical initiation energy not only for hydrogen-air mixtures but for most of the common gaseous hydrocarbon fuels ranging from ethylene to the alkanes. Further correlation requires additional data on the quantitative measurement of initiation energy, the bulk of which so far has been produced by Ellsworth at the Shell Thornton Research Center in Great Britain. The significant importance of the cell size - initiation energy correlation cannot be overemphasized since now via a relatively simple laboratory scale measurement of cell size λ , one can infer the initiation energy and hence shock sensitivity of just about any explosive gaseous medium. Finally, detonability limits have also been correlated in terms of the cell size λ . Apart from the direct importance of being able to deduce detonability limits again from a relatively straightforward procedure of measuring the cell size, there arises the further important conclusion that one cannot define detonability limits in terms of thermochemical properties of the mixture alone but that one must also define the confining geometrical boundary conditions to unambiguously define the true detonability limits for a given mixture in a particular configuration.

Thus in terms of the progress report, Appendix I describes in great detail the recent successful universal quantitative correlation between the cell size λ and the other practically

important "dynamic" detonation parameters such as the critical tube diameter, initiation energy and detonability limits. Appendix II followed-up by large scale studies described in Appendix III and Appendix IV pertain to the assessment of the transmission properties of detonation waves from confined to unconfined geometries. Appendix V constitutes a summary review or compendium of essentially the current state of the art in FAE research derived largely from the studies described herein.

3. Intense Turbulent Mixing, Flame Acceleration and Transition to Detonation

In the FAE III concept, one relies on the tailored intense turbulent mixing between the chemical sensitizer and the explosive fuel-air cloud to set up suitable conditions for "shockless" initiation of detonation. By "shockless" initiation is meant that one does not supply a priori a detonation level shock wave in the initiation process as one does, for example, in the blast initiation. In fact, the medium initially is entirely shock free. Rather the shock wave, which ultimately must reach detonation strength, must evolve spontaneously within the chemically sensitized activation region purely by virtue of the intensity of the exothermic chemical reactions relying exclusively on the energy content of the sensitized portion of the fuel-air cloud itself. Clearly, phenomenologically this is not a simple process and to work successfully requires a very judicious tailoring of a number of time scales, length scales,

reaction rates and energetics. Suitable conditions must be generated such that once reactions are initiated rapid amplification of pressure waves that are produced in the exothermic chemical reactions must progress in a coherent fashion to lead to detonation level shock waves within the chemically sensitized region which must be of sufficient size, the scale of which has been alluded to in the discussion on the critical tube diameter in the previous section. The "detonation" wave thus formed within the chemically sensitized portion of the fuel-air cloud will then transmit into the rest of the cloud leading to global detonation of the cloud. The most difficult aspect of the FAE III concept is to know a priori and be able to create experimentally the kind of conditions whereby the chemical reactions in the sensitized portion of the cloud progresses in step (i.e. coherently) with the evolving pressure field and thereby provide the adequate shock amplification environment which can ultimately lead to detonating the initially shock free medium.

In the current program year, emphasis was placed on the study of the problem of intense turbulent mixing between the burnt and the unburnt explosive mixture to explore specifically the environment under which rapid amplification to detonation can be achieved. The turbulent mixing was induced by having a weakly initiated flame propagate into an obstacle environment along a linear combustion tube and thereby create the intense turbulent flow field ahead of itself by virtue of the fluid

displacement as a result of the increase of the specific volume in an exothermic reaction. The conditions of intense turbulent mixing between the burnt and unburnt FAE mixture, the resulting rapid flame acceleration and the minimum necessary conditions for transition to detonation have been studied in the present program and are reported in Appendix VI.

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APPENDIX I

MEASUREMENTS OF CELL SIZE IN HYDROCARBON-AIR MIXTURES AND PREDICTIONS OF
CRITICAL TUBE DIAMETER, CRITICAL INITIATION ENERGY AND DETONABILITY LIMITS

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ABSTRACT

Experimental measurements of the detonation cell size in mixtures of H_2 , C_2H_2 , C_2H_4 , C_2H_6 , C_3H_8 and C_4H_{10} with air over a range of fuel concentrations have been carried out in three cylindrical tubes of diameters 5 cm, 15 cm and 30 cm. The cell size has been determined from the signatures on smoked aluminum foils placed inside the tube as well as from the frequency of the pressure fluctuations recorded by piezoelectric transducers. Based on the cell size data obtained, estimates of the critical tube diameter using the empirical law of Soloukhin and Mitrofanov ($d_c = 13\lambda$) have been found to be in agreement with experimental data from direct measurement of the critical tube itself. Hence, the important empirical law $d_c = 13\lambda$ is thus verified. Estimates of the critical charge weight from the cell size data using the surface energy theory proposed by Lee have been found to agree reasonably well with the experimental results of Elsworth. Based on the criteria for stable propagation in tubes ($d^* = \lambda/\pi$) and in two-dimensional channels ($W^* = 3\lambda$), detonability limits can also be predicted from a knowledge of the cell size λ . Based on Westbrook's kinetic calculations, it is found that the cell size data are directly proportional to the induction time of the oxidation process which confirms qualitatively Schelkhin's model. However, on a quantitative basis, Schelkhin's model predicts cell sizes an order of magnitude larger than the present experimental data.

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MEASUREMENTS OF CELL SIZE IN HYDROCARBON-AIR MIXTURES AND PREDICTIONS OF CRITICAL TUBE DIAMETER, CRITICAL INITIATION ENERGY AND DETONABILITY LIMITS

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I. Introduction

An assessment of the relative detonation sensitivity of various fuel-oxygen-nitrogen mixtures was carried out by Matsui and Lee¹ using the critical initiation energy for spherical detonations as a basis for comparison. The critical initiation energy itself depends on the energy-time characteristics as well as the geometry of the source^{2,3} and is thus a rather complex parameter. Furthermore, for the relatively insensitive fuel-air mixtures, the critical initiation energy is usually measured in terms of an equivalent weight of a high explosive charge typically of the order of 100 g or more. This requires rather large volumes of gases for a long enough detonation travel to conclusively determine the stability of the detonation in the case of successful initiation. The use of long rectangular bags rather than spherical balloons simplifies the experiments somewhat but suffers from a certain degree of lateral confinement of the plastic walls as shown by Moen et al.⁴ Thus, a rectangular bag does not simulate a truly unconfined spherical detonation. For insensitive fuels or for near-limit mixtures, the critical cross-sectional dimension of the rectangular bag required for stable propagation is also very large and is not known a priori.

In recent years, the critical tube diameter required for the successful transformation of a confined planar detonation into an unconfined spherical wave has been shown to be easily measurable. Since the re-initiation process for successful transformation of a planar into a spherical wave occurs within a distance of the order of one tube diameter from the exit of the tube, even the large-scale experiments required are of rather modest dimensions. Although the critical tube diameter has been related to the critical energy by Lee and Matsui⁵ using a "work-done" concept, a far more important fundamental relationship between the critical tube diameter " d_c " and the cell size or transverse wave spacing " λ " has been established in recent years. First observed by Mitrofanov and Soloukhin⁶ and confirmed later by Edwards et al.,⁷ it was found that $d_c = 13\lambda$ for $C_2H_2-O_2$ mixtures at sub-atmospheric pressures of about 80 torr. Following the suggestion of Edwards that this empirical law should have a wider applicability, Knystautas et al.⁸ carried out extensive experiments in fuel-oxygen-nitrogen mixtures to measure both the critical tube diameter " d_c " as well as the cell size " λ " over a wide range of initial pressures and confirmed the validity of this empirical law of $d_c = 13\lambda$ for the fuels tested (H_2 , C_2H_2 , C_2H_4 , C_3H_6 , C_3H_8 , CH_4 , C_2H_6 , C_4H_{10} and MAPP). With the critical tube diameter linked to the cell size through this simple empirical law, it is then possible to perform laboratory-scale experiments in confined tubes to determine the cell size " λ " and deduce the critical tube diameter from it. A program for the systematic measurement of the detonation cell size in atmospheric fuel-air mixtures has been carried out. The present paper reports the results obtained and the correlation of the cell size to existing data from actual measurements of critical tube diameter as well as critical initiation energies.

2. Experimental Details

Three steel tubes (5, 15 and 30 cm in diameter) of length ≈ 20 m were used in the present study. The tubes were equipped with ionization probes for the measurement of the detonation velocity and piezoelectric transducers for pressure measurements. Commercial grades of C_2H_2 , H_2 , C_2H_4 , C_2H_6 , C_3H_8 and C_4H_{10} and bottled compressed air were used. In the 5 cm and 15 cm diameter tubes, the mixture of the desired composition was prepared in a continuous flow system with flow rates monitored through standard calibrated rotameters. The tube was purged with the premixed gases for at least 5 tube volumes prior to the experiment. In the larger diameter (30 cm) tube, the mixing procedure was as follows: the tube was first evacuated and the desired volumes of air and fuel were then introduced into the tube via the method of partial pressures. A bellows-type pump was then used to recirculate the gases from one end to the other to permit thorough mixing of the components. Initiation of the detonation was via an exploding wire or a blasting cap depending on the sensitivity of the mixture. In the 30 cm diameter tube, it was often required to use an additional booster charge with the blasting cap for direct initiation. A short length of wire spiral was usually placed at the initiation end to guarantee the rapid formation of the detonation wave. To record the detonation cell signatures, smoked aluminum foils were used. The aluminum foils were usually of width πd and as long a length (in the direction of propagation) as experimentally possible for a more easier estimation of the averaged cell size. For cell sizes large compared to the diameter of the pressure transducer, periodic pressure fluctuations could be seen superimposed on the main pressure trace. The periods of these pressure fluctuations " t_c " can be used to estimate the cell length " L_c " ($L_c \approx t_c V_{CJ}$) which can then be converted into cell size via the approximate geometrical relationship $\lambda \approx 0.6 L_c$. Thus the pressure trace as well as the smoked foil record provide two independent means for estimating cell sizes. In general, a few experiments had to be carried out for each mixture composition in order to conclusively establish the cell size for the particular mixture.

2. Results

The averaged detonation cell diameter " λ " for six gaseous fuels (H_2 , C_2H_2 , C_2H_4 , C_2H_6 , C_3H_8 and C_4H_{10}) is plotted against the equivalence ratio ϕ in Fig. 1. Except for hydrogen, the minimum cell size (or the most sensitive composition) corresponds to a mixture composition slightly on the fuel-rich side ($\phi > 1$) rather than stoichiometric ($\phi = 1$). As can be observed, estimates of the cell size from pressure fluctuations are in good agreement with those obtained from smoked foil records. Except for methane (CH_4), all the alkanes (C_3H_8 , C_2H_6 , C_4H_{10}) appear to have the same sensitivity in that their cell sizes are practically identical. For methane, the use of 50 g of explosive charge failed to cause direct initiation for the stoichiometric composition although Kogarko⁹ reported detonations in fuel-lean (from 6.9 to 8.2% CH_4) and fuel-rich (from 11.1 to 13.5% CH_4) CH_4 -air mixtures in a similar tube using 50 g and 70 g of explosive charges, respectively. However, we feel that a 50 g charge is the upper limit for safe operation in a university laboratory. Furthermore, Moen¹⁰ recently performed experiments in a 6 foot diameter tube and estimated from his smoked foil record a cell size of about 33 cm for stoichiometric methane-air mixtures. This would correspond to about 6 times the minimum cell size of about 5.35 cm for the other alkanes (i.e., C_3H_8 , C_2H_6 and C_4H_{10}). In increasing order of sensitivity (or decreasing cell size), ethylene (C_2H_4) follows the alkanes and hydrogen (H_2) is slightly more sensitive than C_2H_4 . As expected, acetylene is found to be the most sensitive fuel with a minimum cell size $\lambda \approx 0.565$ cm as compared to $\lambda \approx 1.5$ cm for H_2 , $\lambda \approx 2.6$ cm for C_2H_4 and $\lambda \approx 5.35$ cm for the alkane group (C_3H_8 , C_2H_6 and C_4H_{10}). For the stoichiometric compositions of

the mixtures studied in the present work, cell sizes were determined by Bull et al.¹¹ in a detonation tube of rectangular cross section (7.6 cm x 3.8 cm) using smoked stainless steel foils. Bull's results are $\lambda \approx 0.92$ cm, 1. cm, 2.4 cm, 5.4 cm and 5.6 cm for C_2H_2 , H_2 , C_2H_4 , C_2H_6 and C_3H_8 , respectively and are in good agreement with the present observations. It is also of importance to note that the width of the typical U-shaped curves for cell size λ versus fuel composition ϕ decreases with decreasing sensitivity of the fuel. Also, the increase in cell size as the equivalence ratio approaches the rich limit ($\phi > 1$) is in general much slower than towards the lean limit ($\phi < 1$). This universal behavior is reflected in practically all dynamic detonation parameters that are dependent on the induction time of the mixture itself.

Based on cell size data, the critical tube diameter d_c can be estimated from the empirical relationship $d_c \approx 13\lambda$. The solid curves shown in Fig. 2 represent critical tube diameters estimated from the present data obtained for λ . Although an extensive experimental measurement of the critical tube diameter for fuel-oxygen-nitrogen mixtures was carried out by Knystautas et al.⁸, relatively few data exist for " d_c " in fuel-air mixtures, in particular for off-stoichiometric compositions. Some of the data that are currently available from large-scale field tests are also shown in Fig. 2. Because of the interest in H_2 -air mixtures in connection with nuclear reactor safety, large-scale experiments on the measurements of d_c for both lean and rich H_2 -air mixtures were carried out recently by Benedick at Sandia National Laboratory (New Mexico).¹² As can be observed from Fig. 2, the agreement with the estimates of d_c from cell size data via the empirical law $d_c \approx 13\lambda$ is extremely good for the case of H_2 . For ethylene (C_2H_4), a limited number of experiments were carried out at Raufoss (Norway)¹³ and at DRES¹⁴ for lean C_2H_4 -air mixtures. The results are also in accord with the present estimates from cell size data. For lean acetylene-air mixtures, some direct measurements of " d_c " were also carried out at Raufoss.¹³ Although no special attempt has been made to narrow down the range of mixture compositions between success and failure for transmission in a given tube, the results are also found to be in good agreement with the predictions from cell size data. For stoichiometric C_2H_2 -air and H_2 -air mixtures, direct measurements of " d_c " were carried out by Knystautas et al.⁸ For stoichiometric H_2 -air mixtures, the agreement is almost perfect. For C_2H_2 -air mixtures, the measurements of " d_c " are slightly higher than those estimated from " λ ". However, "bottle" to "bottle" variations of the purity of the commercial C_2H_2 used can account for the observed deviation in view of the extreme sensitivity of the C_2H_2 -air mixture itself. It would be of interest to extend the critical tube diameter measurements to fuel-rich hydrocarbon-air mixtures as in the case of hydrogen-air mixtures in order to verify the validity of the $d_c \approx 13\lambda$ correlation over the entire range of fuel compositions between the two limits.

Although the critical initiation energy has been deduced from a knowledge of the critical tube diameter from energy considerations using a "work done" concept,^{5,15} a better model was recently developed by Lee.^{12,16} The model postulates that there exists a minimum surface energy per unit area of the wave front for successful transformation of a planar detonation into an unconfined spherical wave. This minimum "surface energy" is then equated directly to the area of the critical tube (i.e., $\pi d_c^2/4$). Assuming the requirement of a minimum size of a detonation kernel for direct initiation,¹⁷ the surface area of the critical detonation kernel is then equated to the area of the critical tube (i.e., $4\pi R_k^2 = \pi d_c^2/4$). Using strong blast theory to relate the blast energy E_c to the kernel radius R_k where the detonation is assumed to be of Chapman-Jouguet strength, the following simple expression

was obtained by Lee et al.¹⁶

$$E_c = 4\pi\gamma_0 p_0 M_{CJ}^2 I \left(\frac{d_c}{4}\right)^3 \quad (1)$$

or equivalently

$$E_c = \frac{2197}{16} \pi\gamma_0 p_0 M_{CJ}^2 I \lambda^3 \quad (2)$$

where use has been made of the empirical relationship discussed previously that $d_c \approx 13\lambda$. In Eqs. 1 and 2, γ_0 and p_0 denote the specific heat ratio and the initial pressure of the mixture, M_{CJ} is the Chapman-Jouquet detonation Mach No. and I is a dimensionless constant representing the energy integral in strong blast theory. Based on the cell size data obtained, the critical initiation energies for the various mixtures studied can be evaluated from Eq. 2. Figure 3 shows a comparison of the predicted results using Eq. 2 and those obtained by Elsworth from direct experimental measurements of the critical weight. For C_2H_4 , Murray et al.⁴ have also obtained a few critical initiation charge weights for lean mixtures. The critical energy in Fig. 3 has been expressed as an equivalent charge weight of tetryl (1 g tetryl is equivalent to 4270 joules) for easy comparison with Elsworth's experimental data. The agreement in general is reasonably good in view of the simplicity of the model. No experimental data of critical energies have been reported for C_2H_2 -air mixtures to permit a comparison with the present prediction from Eq. 2. For most practical situations, Eq. 2 can be used to predict critical initiation energies (or charge weights) with quite acceptable accuracy.

The detonability limits in tubes have also been related directly to the cell size by Lee et al.¹⁶ Based on the experimental work of Donato,^{18,19} it was found that the onset of single-headed spinning detonations correspond to the limit for stable propagation in the given tube. Donato showed that for mixtures outside the single-head limit, a finite perturbation would cause the detonation to fail and the combustion wave will continue to propagate as a deflagration thereafter. Only for mixtures within the single-head limit would regeneration into the detonation mode occur shortly downstream of the finite perturbation that causes the wave to fail. It should be emphasized that using a strong enough initiator, spinning detonations can always be initiated in a given tube over a wide range of composition beyond the onset limit when spinning first occurs. However, it has been shown that these spinning waves will decay when encountering finite perturbations and after failure will not transit again to the detonation mode without the support of a strong initiator. However, without a finite perturbation (such as a couple of turns of a Schelkhin spiral), these spinning waves may persist for as much as 100 tube diameters without showing any sign of decay.

Adopting the criterion that the onset of single-head spinning waves in a given tube should correspond to the detonability limits for that tube, the limits in circular tubes can be estimated when cell size data are available. Since the onset of single-head spin corresponds to a cell size of the order of the tube circumference (i.e., $\lambda \approx \pi d$), the composition limits for a given circular tube can be estimated if λ is known. Figure 4 gives the limiting tube diameter " d^* " as function of equivalence ratio for the various fuels studied. For a given fuel composition (i.e., a given ϕ), stable detonations cannot be propagated in tubes with a diameter $d < d^*$. Thus, for the case of stoichiometric H_2 -air mixtures, $d^* \approx 0.5$ cm when compared to $d^* \approx 0.2$ cm for C_2H_2 and $d^* \approx 1.7$ cm for the alkanes. It is of interest to note that according to the cell size reported by Moen for methane (i.e., $\lambda \approx 33$ cm), a minimum tube diameter for stable detonation propagation in stoichiometric methane-air mixtures would be of the order of at least 10 cm. Thus the results

reported by Wolanski et al.²⁰ in a 5 cm tube should correspond to overdriven transient waves only. This is supported by the fact that they observed single-head spinning waves over the whole range of fuel concentrations studied. For two-dimensional channels, the limit should correspond to a composition where the channel height "W" is of the order of the cell size. This is supported by the recent experiments of Vasiliev²¹ who found that stable detonation propagation corresponds to $W \gtrsim \lambda$.

From a practical point of view, the detonation limits for a thin horizontal layer of explosive mixture bounded by one solid surface (i.e., ground) is of interest. Recent experiments by Liu²² for transmission of planar detonations through rectangular slits into unconfined media as well as large scale transmission experiments from two-dimensional channels by Benedick²³ showed that the critical channel width $W^* \approx 3\lambda$. Thus it is reasonable to assume that the minimum thickness "h" of a layer of explosive gas that can support a detonation should be of the order of one and a half cell size (i.e., $h^* \approx 1.5\lambda$). Thus, for stoichiometric H_2 -air mixtures, $h^* \approx 3$ cm as compared to the alkanes C_3H_8 , C_2H_6 and C_4H_{10} for which $h^* \approx 8$ cm. For CH_4 , $h^* \approx 0.5$ m based on Moen's estimate of $\lambda \approx 0.33$ cm, which is much smaller than commonly expected for methane. Direct experiments must be performed to verify the limit postulate of an unconfined thin layer of explosive.

As suggested by Schelkhin,²⁴ the cell size should be directly proportional to the induction zone thickness. However, no theory exists at present whereby the cell size λ can be predicted a priori from a knowledge of the induction kinetics. However, Westbrook²⁵⁻²⁷ has made extensive correlations between experimentally measured λ (or equivalently d_c) and induction times computed from detailed kinetics of the oxidation processes. In general, it can be shown that $\lambda \sim \ell$ where ℓ is the induction length but the constant of proportionality has to be determined by matching with an experimental datum. The proportionality constant depends on how the induction time is computed. For example, Westbrook based his induction time on a constant volume explosion process, while Shepherd²⁸ integrated the kinetic rate equations along a Rayleigh line as in the classical ZDN model for the detonation structure. There is no special justification for either method since both have to use an experimental point to find the proportionality constant itself. The solid curves in Fig. 1 represent the estimated cell sizes from kinetic data matching at the stoichiometric composition point. The results are relatively in good agreement with the data despite some discrepancies as the mixture composition departs from the stoichiometric composition which may result from the choice of the matching point. Nevertheless, the reasonable agreement confirms Schelkhin's postulate. However, useful models for linking the cell size λ to the kinetics of the reaction must somehow take into consideration the complex transient gasdynamics processes of the cell interactions.

4. Conclusions

From the cell size data obtained in the present investigation, critical tube diameter, critical initiation energy (charge weight) as well as detonability limits have been estimated. Comparison with available data for the critical tube from direct experimental measurements confirms the general validity of the empirical law $d_c \approx 13\lambda$ first observed by Mitrofanov and Soloukhin. However, more extensive experiments should be carried out particularly in fuel-rich mixtures to consolidate the validity of this empirical expression. Due to its fundamental significance, it is also of importance to carry out experiments in fuels other than the hydrocarbons to verify the universality of the $d_c \approx 13\lambda$ law. The critical initiation energies predicted by the minimum surface energy model using experimental cell size data agree

reasonably well with direct experimental measurements. Thus, apart from the practical usefulness of Eq. 2, the linkage of the cell size λ to the critical initiation energy E_c should elucidate the fundamental mechanisms of the failure and reinitiating processes in the critical tube experiment, the blast wave initiation processes as well as the detailed cell dynamics of the transient shock interactions. This may lead to a theory for the empirical law $d_c \approx 13\lambda$ itself. Prediction of detonability limits (or minimum tube diameters for stable propagation) from cell size data is an important contribution. However, extensive experiments must be carried out further to verify the postulates for stable propagation (i.e., $d^* \approx \lambda/\pi$ for circular tubes, $W^* \approx 3\lambda$ for two-dimensional channels and $h^* \approx 1.5\lambda$ for a thin layer bounded by one solid surface). Thus far, the experiments in circular tubes by Donato and two-dimensional channels by Vasiliev are of limited scope. Although it has been shown that all the dynamic detonation parameters (i.e., critical tube diameter, critical initiation energy and detonability limits) can be linked to the cell size, a theory for the prediction of the cell size λ from basic information on the physical chemical properties of the explosive mixture is still lacking.

5. Acknowledgement

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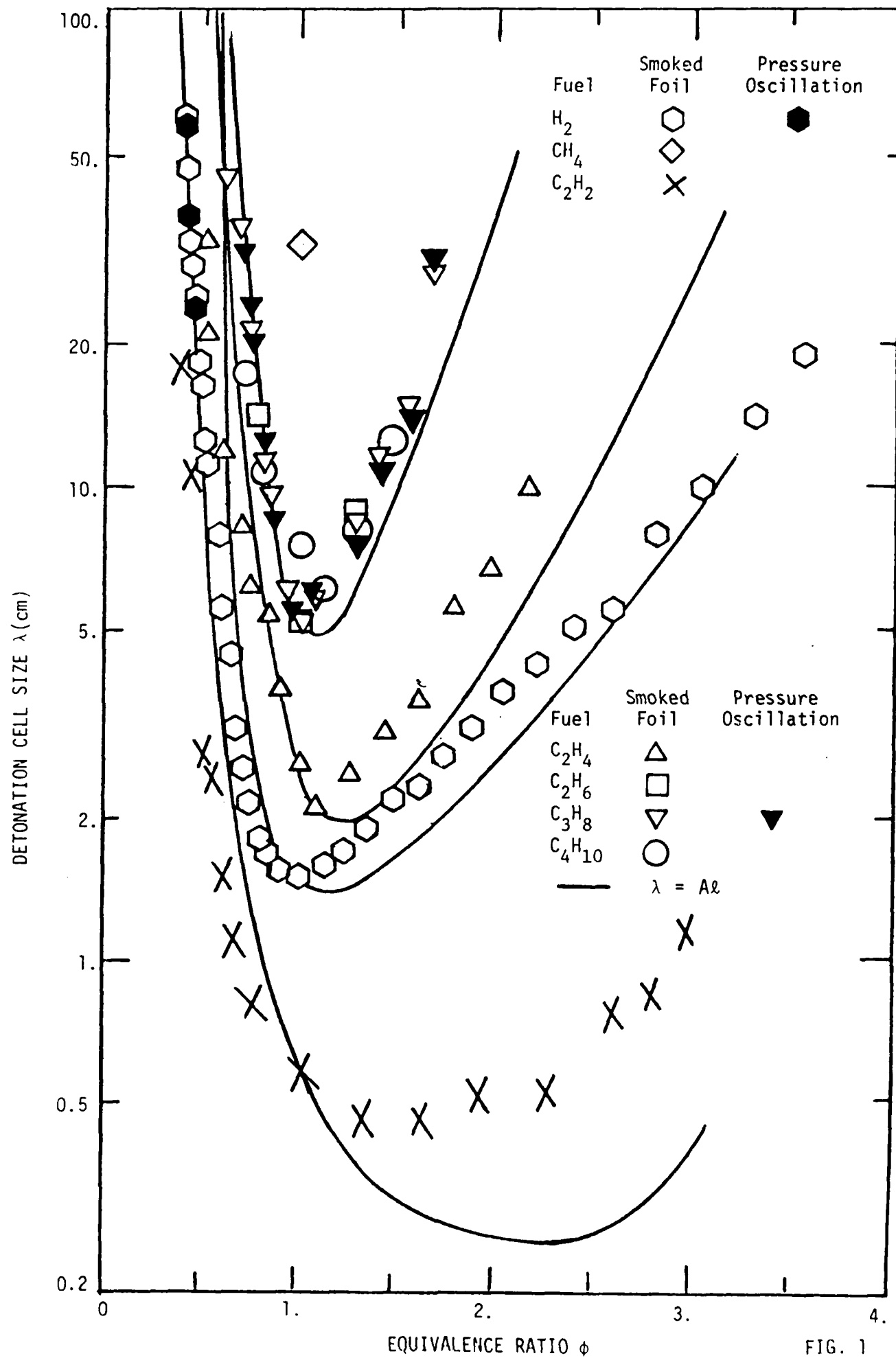


FIG. 1

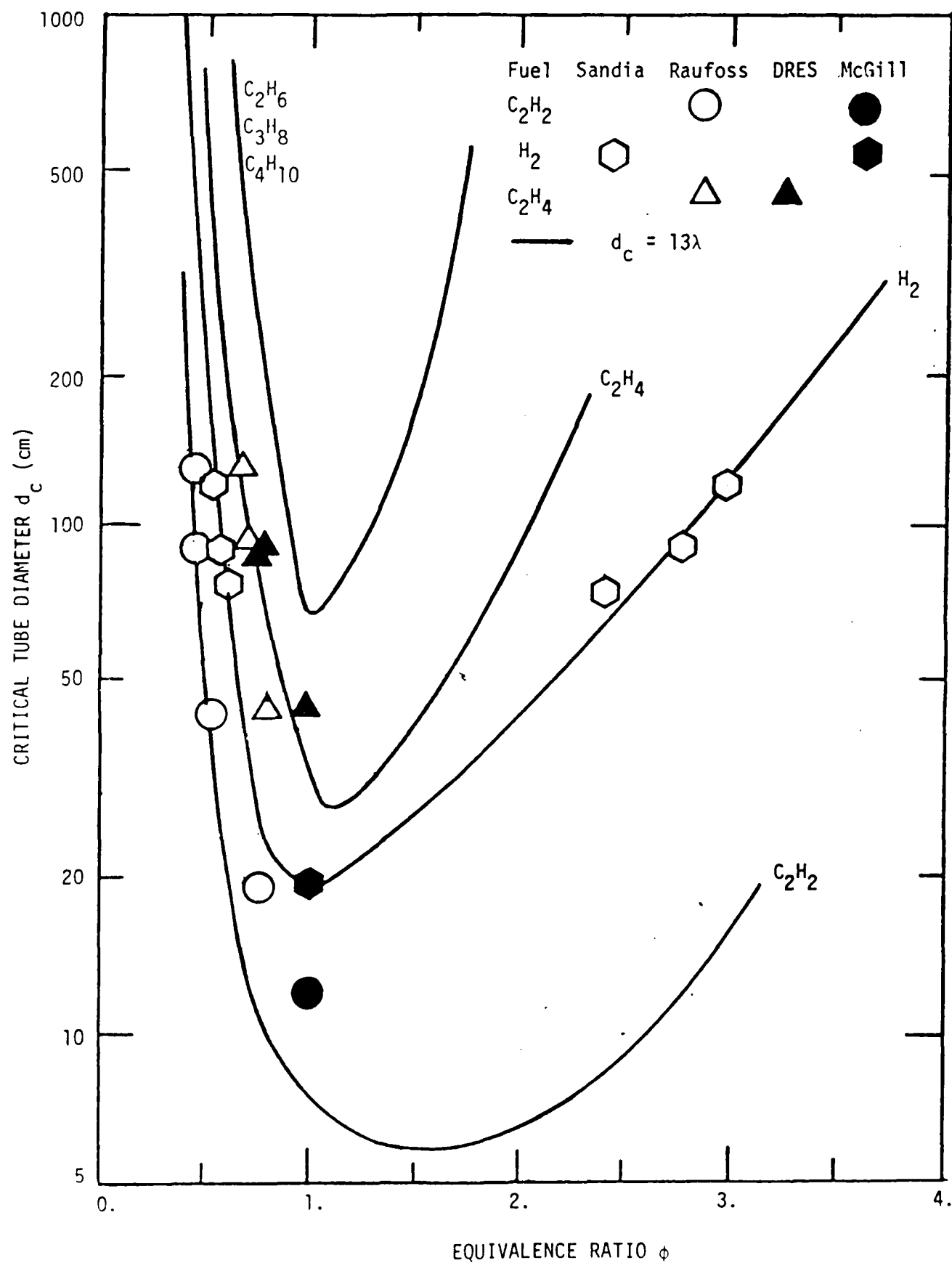


FIG. 2

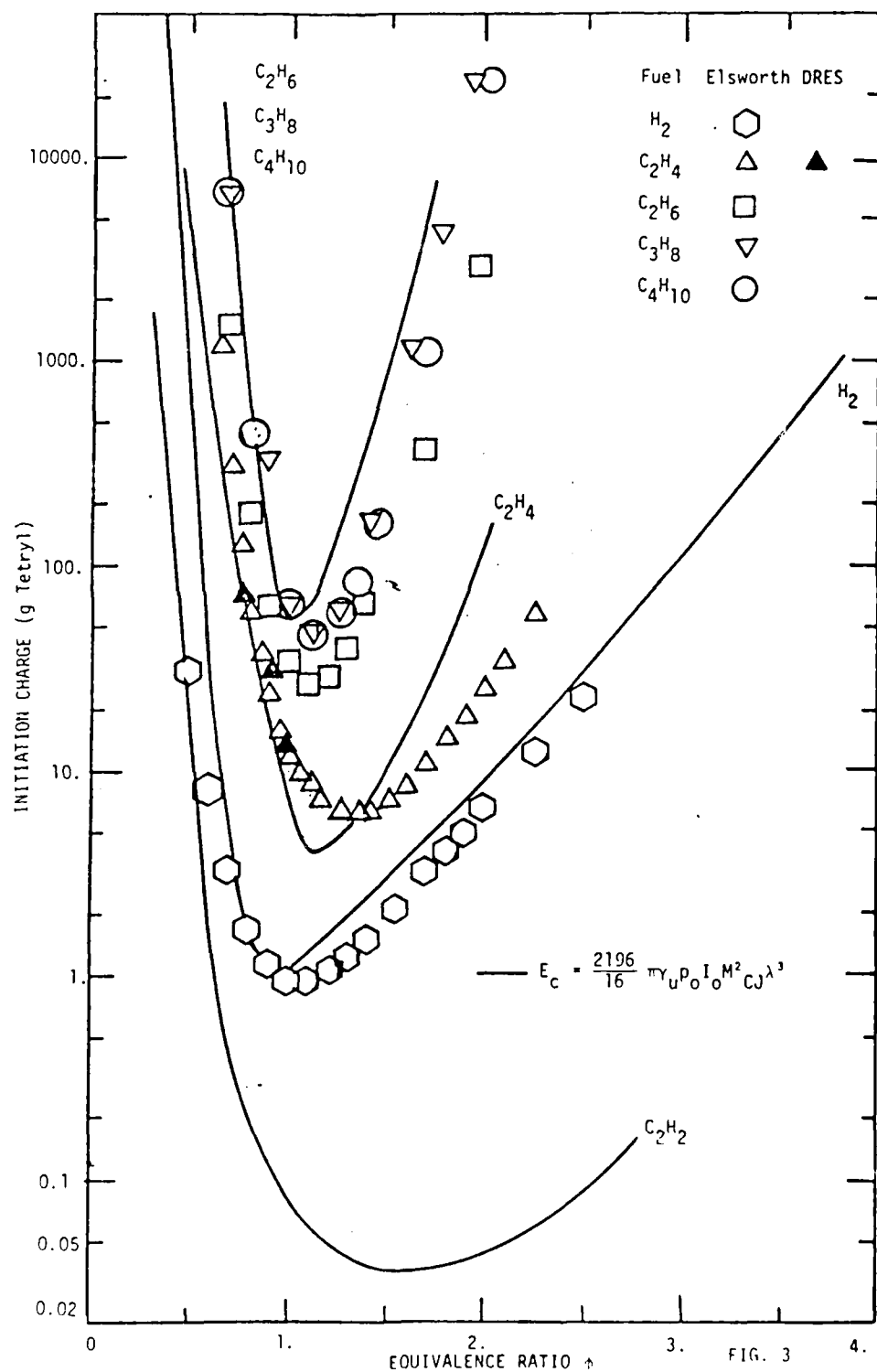


FIG. 3

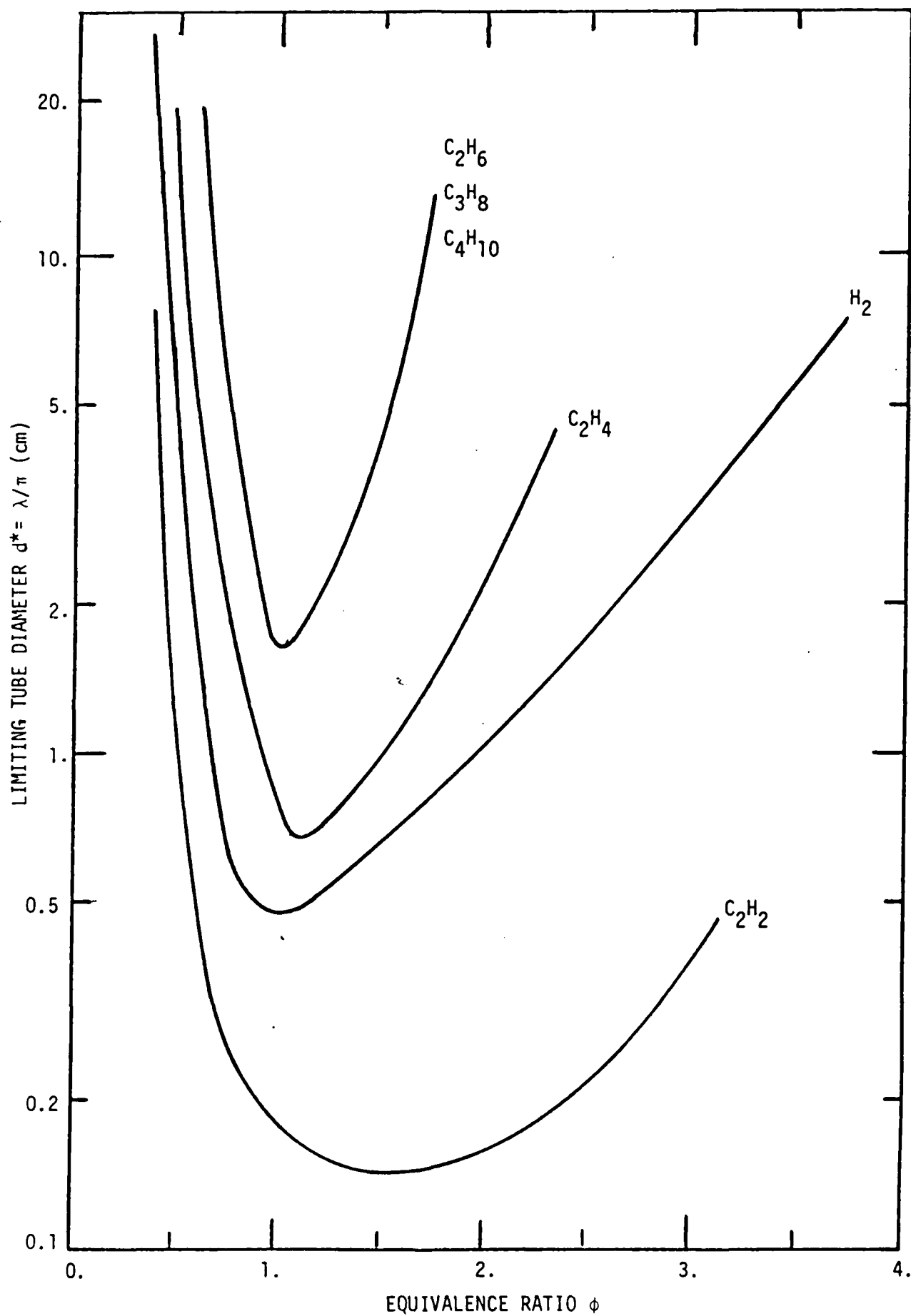


FIG. 4

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4. TITLE (and Subtitle) MEASUREMENTS OF CELL SIZE IN HYDROCARBON-AIR MIXTURES AND PREDICTIONS OF CRITICAL TUBE DIAMETER' CRITICAL INITIATION ENERGY AND DETONABILITY LIMITS		5. TYPE OF REPORT & PERIOD COVERED FINAL PROGRESS May 1 1982-April 30 1983
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Experimental measurements of the detonation cell size in mixtures of H_2 , C_2H_2 , C_2H_4 , C_2H_6 , C_3H_8 and C_4H_{10} with air over a range of fuel concentrations have been carried out in three cylindrical tubes of diameters 5 cm, 15 cm and 30 cm. The cell size has been determined from the signatures on smoked aluminum foils placed inside the tube as well as from the frequency of the pressure fluctuations recorded by piezoelectric transducers. Based on the cell size data obtained, estimates of the critical tube diameter using the		

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empirical law of Soloukhin and Mitrofanov ($d_c \approx 13\lambda$) have been found to be in agreement with experimental data from direct measurement of the critical tube itself. Hence, the important empirical law $d_c \approx 13\lambda$ is thus verified. Estimates of the critical charge weight from the cell size data using the surface energy theory proposed by Lee have been found to agree reasonably well with the experimental results of Elsworth. Based on the criteria for stable propagation in tubes ($d^* \approx \lambda/\pi$) and in two-dimensional channels ($W^* \approx \lambda$), detonability limits can also be predicted from a knowledge of the cell size λ . Based on Westbrook's kinetic calculations, it is found that the cell size data are directly proportional to the induction time of the oxidation process which confirms qualitatively Schelkhin's model. However, on a quantitative basis, Schelkhin's model predicts cell sizes an order of magnitude larger than the present experimental data.

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APPENDIX II

Effect of Geometry on the Transmission of Detonation through an Orifice

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An experimental study has been carried out to investigate the transmission of a planar detonation wave through an orifice into an unconfined medium. Mixtures of $2\text{H}_2 + \text{O}_2 + \beta\text{N}_2$ and $\text{C}_2\text{H}_4 + 3(\text{O}_2 + \beta\text{N}_2)$ for a range of nitrogen concentrations corresponding to $1 \leq \beta \leq 3.76$ and at an initial pressure of 1 atm were used in the experiments. It is found that the critical diameter for the transmission through an orifice is identical to that for a straight tube and both follow the empirical correlation of $d_c \approx 13\lambda$. The transmission through square, triangular, elliptical, and rectangular orifices has led to the development of a correlation based on the effective diameter similar to the case of circular geometry (i.e., $d_{eff} \approx 13\lambda$). The effective diameter is defined as the mean value of the longest and shortest dimensions of the orifice shape. The effective diameter correlation suggests that the criterion for transmission may be based on the mean curvature of the wave front, the implication being that it is not to exceed a certain critical value. The results suggest that local properties in the immediate vicinity of the wave front are the controlling parameters for reinitiation rather than the properties of the gas dynamic flow structure in the wake. Expressions are developed to provide estimates for the critical transmission dimensions for arbitrarily shaped openings. For the two-dimensional limit when one of the characteristic linear dimensions becomes very large compared to the other, it is found that the transmission is based on a critical value for the shorter dimension of the order of 3 times the cell diameter. This result is in accordance with the recent large scale experiments of Benedick in two-dimensional channels of an aspect ratio L/W as large as 35. These observed results can also be successfully explained in terms of the critical wave curvature criterion.

1. INTRODUCTION

When a planar detonation wave emerges from a circular tube into an unconfined environment filled with the same explosive gas mixture, the detonation may continuously transform to propagate as a spherical wave without failure if the tube diameter is greater than a critical value ($d > d_c$) or may fail to a deflagration wave if $d < d_c$. Zeldovich et al. [1] were the first to suggest that the critical diameter is related to the induction zone length of the detonation wave. Mitrofanov and Soloukhin [2] later made the important observation that the critical tube diameter $d_c \approx 13\lambda$ for

a circular tube and $W_c = 10\lambda$ for a rectangular channel (W_c is the channel width and λ is the cell size of the detonation wave). This observation was later confirmed by Edwards et al. [3], who also suggested that the correlation may be universal and valid for all mixtures. The empirical correlation $d_c = 13\lambda$ gives a direct link between the detonation transmission phenomena and the microstructure of the detonation front itself. By using this correlation, other dynamic detonation properties of mixtures (such as the critical initiation energy and detonability limits) may be deduced [4-7]. On the practical side, $d_c = 13\lambda$ is also useful in the design of detonation wave traps and in estimating the critical dimensions of pipes and openings between interconnecting chambers to prevent transmission of detonation from one to the other. Recently, considerable effort has been

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mercial purity (CP grade). The explosive mixture consists of stoichiometric proportions of fuel-oxygen with nitrogen dilution as specified by the parameter $\beta = N_2/O_2$. The range for β is $1 < \beta < 3.76$, the upper bound corresponding to the fuel-air composition. All experiments were carried out at 1 atm initial pressure.

To study the geometric effect of the detonation transmission phenomenon, three groups of orifices, each group corresponding to the same orifice area, were used. Figure 2 illustrates the orifice configurations for each group and consists of circular, square, triangular, and elliptical (2:1 axis ratio) orifices. The three groups of orifices have areas of 20, 64, and 127 cm², corresponding to circular orifice dimensions of 5, 9, and 12.6 cm diam. In order to study the detonation transmission process for a two-dimensional expansion, a series of rectangular orifices was used. The range of aspect ratio (L/W) for the rectangular orifices is $3.75 < L/W < 15$, with the range of actual dimensions being $8 < L < 19$ cm and $1 < W < 5$ cm. To assess the detonation transmission from the detonation tube through the orifice and into the detonation chamber, two calibrated piezoelectric pressure transducers (PCB113A24, 5 mv/psi) were used, one in the detonation tube and the other at the center of the back flange (opposite to the detonation tube exit) of the detonation chamber. Successful transmission into the chamber or failure of the detonation wave in the chamber was established by the time of arrival and the magnitude of the

reflected pressure as monitored by the transducer at the back flange of the chamber. A smoked foil, placed along the inner wall of the 20 cm diam detonation tube, was used to determine the characteristic cell diameter λ for each mixture.

3. RESULTS AND DISCUSSIONS

The preliminary results reported earlier [7] have indicated that the critical diameter for detonation transmission from a tube and an orifice is the same under similar conditions. However, due to a higher upstream pressure from the reflection of the detonation wave from the orifice plate, one may expect that the critical orifice diameter should be somewhat less than that for a straight tube. Since the results reported earlier are based on a limited number of large scale experiments under field conditions where experimental parameters could not be controlled accurately, a series of test were carried out in $H_2-O_2-N_2$ and $C_2H_4-O_2-N_2$ mixtures at an initial pressure of 1 atm to verify this important result. Figures 3 and 4 show the critical orifice diameter for a range of nitrogen concentration in stoichiometric mixtures [i.e., $2H_2 + O_2 + \beta N_2$ with $1 < \beta < 3.76$ and $C_2H_4 + 3(O_2 + \beta N_2)$ for $1 < \beta < 3.0$]. The results are found to be in perfect accord with those for straight circular tubes obtained previously by Knystautas et al. [4]. Cell size measurements have also been carried out, and the cell size data are also in agreement with those of Knystautas et al. Based on the cell size data, the empirical correlation of $d_c \cong 13\lambda$ is also found to be valid for circular orifices as it is for straight circular tubes. Thus the present results conclusively demonstrate the equivalence of the critical diameter for orifices and for tubes, even though in certain experiments, the ratio of the orifice diameter to the tube diameter in which the orifice plate is mounted is quite small ($d/D \cong 0.1$). One would expect significant influence of the reflection of the detonation wave on the flow structure of the product gases emitting from the orifice under this condition. Thus the present experiments indicate that the reinitiation process as the detonation wave exits from the orifice (or tube) is a very local phenomenon limited to a small region of the order of a cell length in thickness at the detonation front itself.

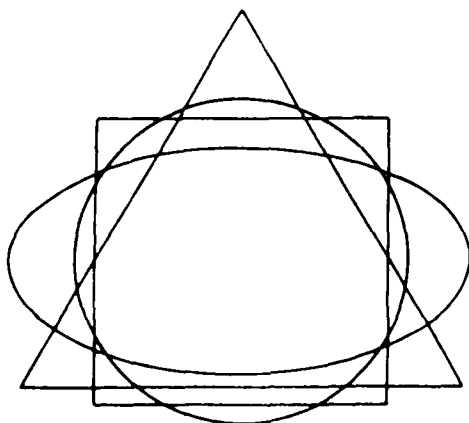


Fig. 2. Comparison of different orifice geometries for the same area.

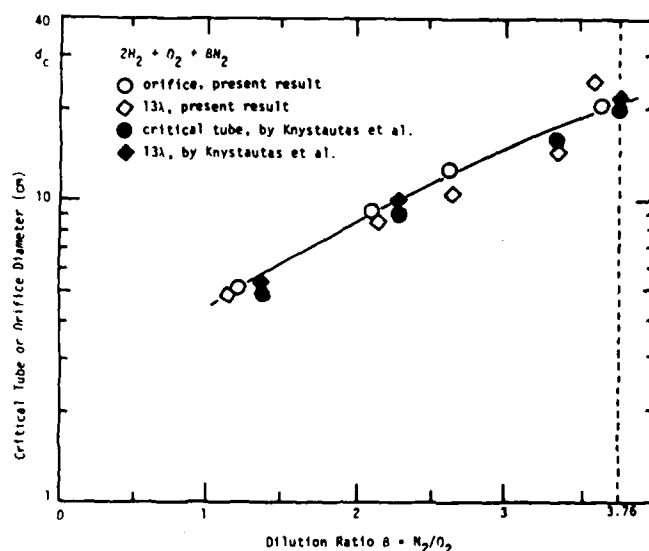


Fig. 3. Comparison of the critical tube and orifice diameters as a function of dilution ratio in H_2 - O_2 - N_2 mixtures.

The flow behind the front seems to play a minor role in the reinitiation process. Hence, the so-called work-done model suggested by Lee and Matsui [8] in which the reinitiation is equated to the blast initiation process, with the blast energy

being equated to the work done by the product gases emitted from the tube (or orifice), does not appear to give the correct description. However, the later surface energy model proposed by Lee [5], where a minimum surface energy is postu-

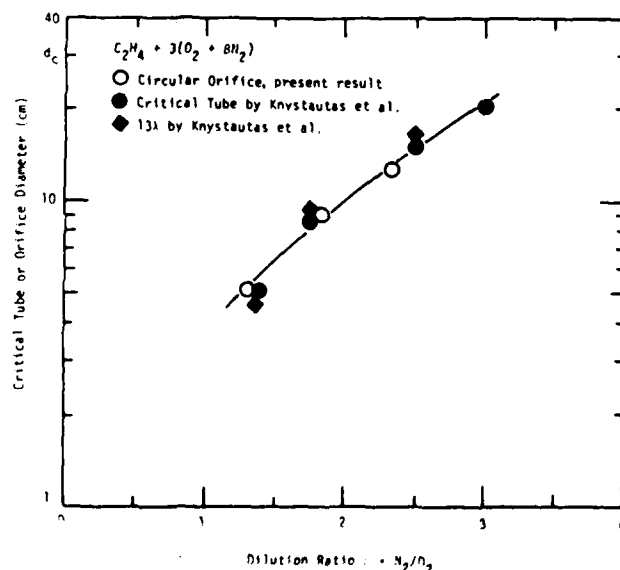


Fig. 4. Comparison of the critical tube and orifice diameters as a function of dilution ratio in C_2H_4 - O_2 - N_2 mixtures.

lated to be required for the evolution of a planar to a spherical wave, appears to be more compatible with the present findings.

For the transmission of detonation through orifices with shapes other than circular, a series of experiments has been carried out for square, triangular, and elliptical orifices for the two mixtures $2\text{H}_2 + \text{O}_2 + \beta\text{N}_2$ for $1 < \beta \leq 3.76$ and $\text{C}_2\text{H}_4 + 3(\text{O}_2 + \beta\text{N}_2)$ for $1 < \beta \leq 3$ at initial pressures of 1 atm. Attempts to use the hydraulic diameter ($d_H = 4 \times \text{area/wetted perimeter}$) to characterize the different geometrical openings and seek a similar correlation between the critical hydraulic diameter and the cell size of the mixture as was done in the case of the circular geometry (i.e., $d_c = 13\lambda$) failed to yield meaningful results. Figures 5 and 6 give the critical hydraulic diameter d_H for the H_2 and C_2H_4 mixtures for various values of nitrogen concentration for the square, triangular, and elliptical orifices. Each group of points corresponds to the same orifice area. As can be observed, for the same orifice area, the circular orifice gives the largest critical hydraulic diameter, or for the same hydraulic diameter, the circular orifice requires the most sensitive mixture (i.e., the smallest value of β). The triangular orifice appears to be the most effective in the transmis-

sion of the detonation for the same value of the hydraulic diameter, being distinctly more effective than the square or the elliptical orifices. This suggests that the critical hydraulic diameter is geometry dependent. No universal correlation is observed for the differently shaped orifices, and the hydraulic diameter does not appear to be the appropriate correlation parameter to describe the detonation transmission process. However, it is found that one can define an effective diameter d_{eff} based on the mean value of diameter of the inscribed d_1 and the circumscribed d_2 circles for each of the geometrical openings [i.e., $d_{eff} = (d_1 + d_2)/2$], and this effective diameter appears to follow the same universal empirical law of $d_{eff} = 13\lambda$ as for the case of the circular orifice geometry. The results for d_{eff}/λ plotted against the hydraulic diameter for the three geometries (i.e., square, triangular, and elliptical orifices) for both H_2 and C_2H_4 mixtures are given in Figs. 7-9. As indicated in these figures, the experimental results show that $d_{eff} \approx 13\lambda$ for all the geometries. That the effective diameter $d_{eff} = (d_1 + d_2)/2$ is found to be equivalent to the diameter of a circular orifice suggests that the reinitiation process depends on the curvature of the wave front. Upon emerging from the orifice

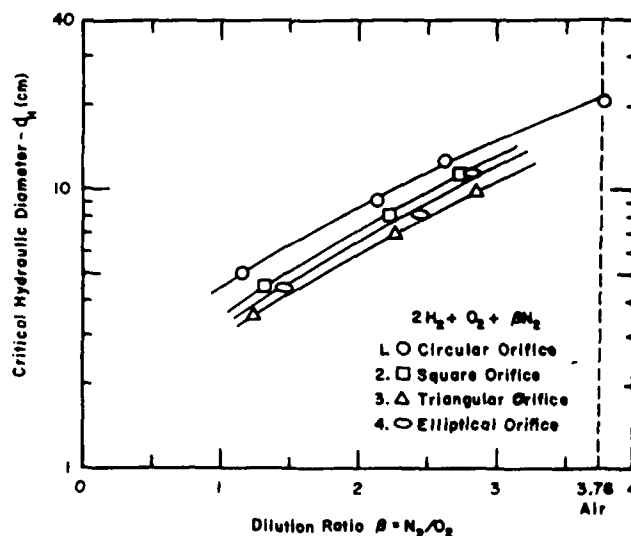


Fig. 5. Variation of the critical hydraulic diameter with dilution ratio in $\text{H}_2\text{-O}_2\text{-N}_2$ mixtures.

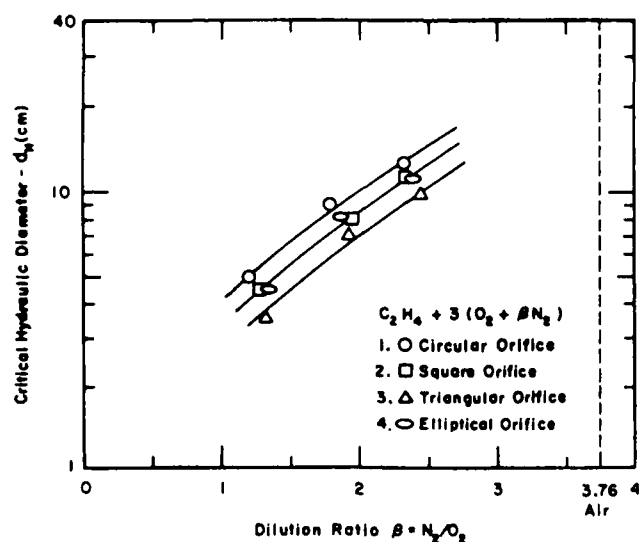


Fig. 6. Variation of the critical hydraulic diameter with dilution ratio in C_2H_4 - O_2 - N_2 mixtures.

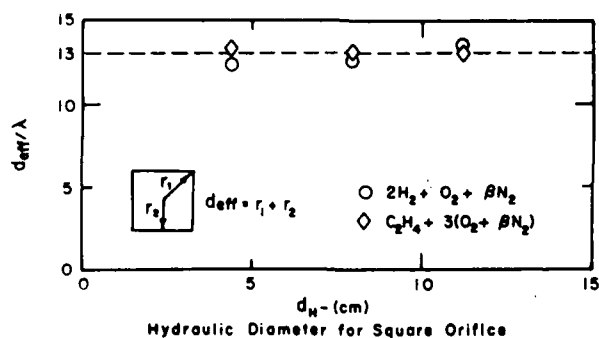


Fig. 7. Variation of the effective diameter with hydraulic diameter for the square orifice.

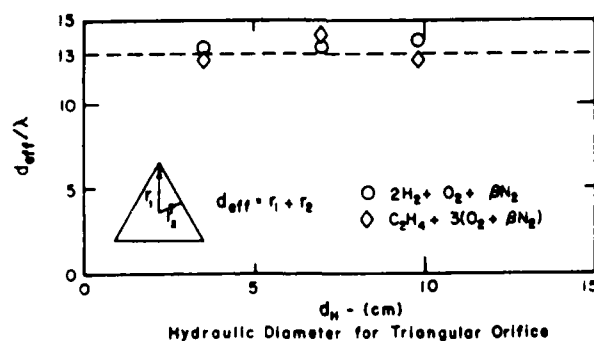


Fig. 8. Variation of the effective diameter with hydraulic diameter for the triangular orifice.

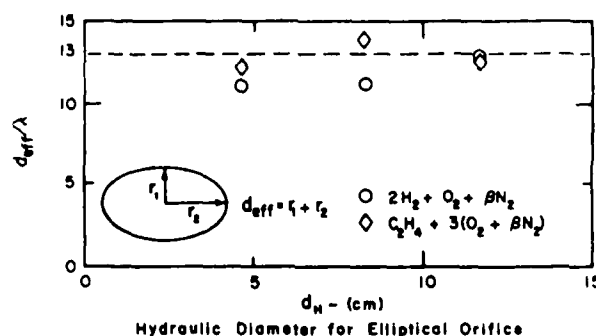


Fig. 9. Variation of the effective diameter with hydraulic diameter for the elliptical orifice.

(or tube), the planar detonation wave front is subjected to an expansion process. The penetration of the rarefaction waves toward the axis of the detonation gives rise to a curved front as the transformation to an eventual unconfined spherical detonation takes place. The radius of curvature depends on the linear dimension of the front as well as on the geometry. For a circular orifice, there is only one linear dimension (i.e., the tube or orifice diameter), and the existence of a critical diameter indicates a corresponding critical radius of curvature for successful propagation of the curved front. For noncircular geometries, there exist two linear dimensions characterizing the initial wave front shape. The effective diameter being the average of the inscribed and circumscribed circles gives the appropriate length scale on which the mean radius of curvature of the curved detonation front depends. Thus one may expect a similar correlation of $d_{eff} = 13\lambda$ if a certain critical radius of curvature is required for the successful transformation of an eventual spherical wave. The results for different geometries and the successful correlation of $d_{eff} = 13\lambda$ as explained above are compatible with the conclusions stated earlier that the reinitiation process is a very local phenomenon, limited to the region in the immediate vicinity of the detonation front, with little dependence on the flow structure of the product gases emitting from the opening or tube. The key parameter that characterizes the wave front appears to be the mean curvature, which depends strongly on the initial geometry of the detonation front as it exits the orifice and is

subjected to transverse penetration by the rarefaction waves.

Generalizing the above discussions to arbitrary geometries, one may consider, for example, a polygonally shaped orifice of n sides, with l being the length of one side. The hydraulic diameter d_H for the polygon is thus (Fig. 10)

$$d_H = \frac{4A}{nl} = l \cot \frac{\pi}{n} \quad (1)$$

The relation between d_{eff} and the hydraulic diam-

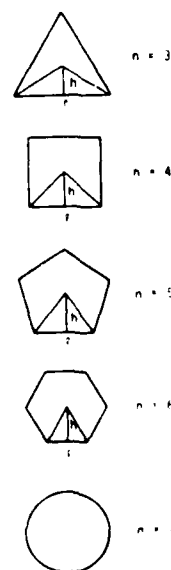


Fig. 10. Schematic illustration of polygon geometry.

eter d_H is therefore

$$d_{eff} = \frac{1 + \cos(\pi/n)}{2 \cos(\pi/n)} d_H. \quad (2)$$

Thus if we use the present experimental results obtained for $d_{eff} = 13\lambda$, the critical hydraulic diameter for the polygonal shaped orifice becomes

$$d_H = \frac{2 \cos(\pi/n)}{1 + \cos(\pi/n)} 13\lambda. \quad (3)$$

For $n \rightarrow \infty$ the above expression yields $d_H \rightarrow 13\lambda$ for the circular orifice. The general expression represented by Eq. (3) can now be used to correlate the experimental data for the square ($n = 4$) and the triangular ($n = 3$) orifices. Table 1 gives a comparison between the prediction given by Eq. (3) and the experimental results. The agreement is quite good, and it would be of interest to extend the experiments to pentagonally and hexagonally shaped orifices to test the general validity of Eq. (3).

For the ellipse, the effective diameter is related to the hydraulic diameter by the expression

$$d_{eff} = \frac{1}{4} (1 + K) \left(1 + \frac{1}{K} \right) \left(\frac{64 - 3B^4}{64 - 16B^2} \right) d_H, \quad (4)$$

where $K = a/b$ is the ratio of the major (a) to the minor (b) axes and $B = (K - 1)/(K + 1)$. Thus using the correlation $d_{eff} = 13\lambda$, we obtain for the critical hydraulic diameter for the ellipse

$$d_H = \frac{52\lambda}{(1 + K)(1 + 1/K)[(64 - 3B^4)/(64 - 16B^2)]} \quad (5)$$

TABLE 1

n	Configuration	Eq. (3)	Experimental Results
3	Triangle	$d_H = 8.66\lambda$	$d_H = 9.02\lambda$
4	Square	$d_H = 10.8\lambda$	$d_H = 10.8\lambda$
5	Pentagon	$d_H = 11.6\lambda$	—
∞	Circle	$d_H = d_c = 13\lambda$	$d_c = 13\lambda$

For $a = b$ (a circular orifice), $K = 1$, and $B = 0$, the above expression yields $d_H = d_c = 13\lambda$. Table 2 compares the predicted results using Eq. (5) with experimental data. The agreement is fair and shows at least the correct qualitative trend of a decreasing d_H as K increases. Again the general validity of Eq. (5) requires further experimental work using elliptical orifices with different values of K .

Rectangular orifices of different aspect ratios (L/W = length of slot/width of slot) are of particular interest since the variation of L/W from one to infinity gives the continuous transition from three- to two-dimensional geometry, corresponding to the detonation evolving to a cylindrical wave. Previous experiments by both Mitrofanov and Soloukhin [2] and Edwards et al. [3] have reported that for square channels $W_c \approx 10\lambda$. This agrees with the present results for the square orifice, where the critical width W_c is found to be in the range $10\lambda < W_c \leq 11\lambda$ for both the H_2 and C_2H_4 mixtures tested. Results for W_c/λ for values of L/W ranging from 1 to 15 are shown in Fig. 11. Also plotted in Fig. 11 are the earlier results of Bjerketvedt et al. [7], who have performed a few experiments with both C_2H_2 and C_2H_4 mixtures for orifices with $1 \leq L/W \leq 5$. The results show that W_c/λ decreases from a value of the order of 10 for $L/W \approx 1$ quite rapidly to an asymptotic value of about 3 for $L/W \geq 7$. In other words, as one approaches the two-dimensional geometry ($L/W \rightarrow \infty$), the critical channel width $W_c \rightarrow 3\lambda$. This rather surprising observation has prompted Benedick et al. [9] to perform some large scale field tests in which two-dimensional channels (instead of rectangular slots) of very large aspect ratios ($L/W \rightarrow 35$) could be used. The recent results obtained by Benedick for H_2 -air mixtures are also plotted in Fig. 11, which confirms the

TABLE 2

K	Configuration	Eq. (5)	Experimental Data
1	Circle	$d_H = d_c = 13\lambda$	$d_c = 13\lambda$
2	Ellipse	$d_H = 11.2\lambda$	$d_H = 10.6$
3	Ellipse	$d_H = 9.2\lambda$	—

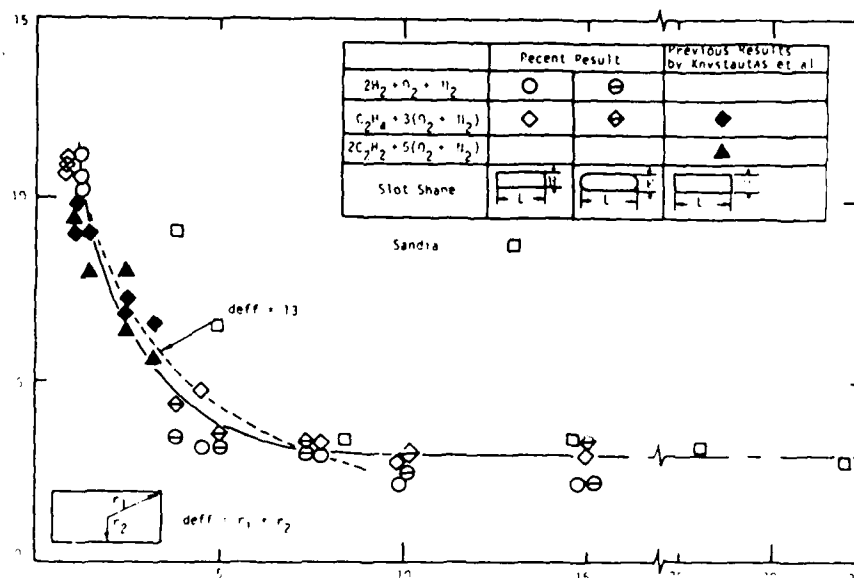


Fig. 11. Variation of the critical slot width with aspect ratio for rectangular orifices.

asymptotic limit value of $W_c/\lambda \approx 3$ that has been obtained in the present study using slot orifices.

To attempt to explain all these observed detonation transmission phenomena, and especially the surprising result that the critical transmission criterion for rectangular orifices changes from $W_c/\lambda \approx 10$ for an aspect ratio $L/W = 1$ (square orifice) to $W_c/\lambda \approx 3$ for aspect ratios $L/W > 7$, one can invoke the recently proposed wave curvature concept of Lee [10]. In the general transmission situation, Lee suggests that when a planar detonation wave propagating in a confining linear tube or channel emerges suddenly into an unconfined volume, it is subject to lateral relief due to the centered expansion wave at the edge of the opening. The rarefaction waves that are thus generated penetrate into the wave front, retard the affected portions, and thereby essentially impose a curvature on the wave front. He then defines the critical condition for transmission in terms of a minimum local radius of curvature of the diffracted wave, namely: If the rarefaction waves give rise to a curvature κ of the diffracted detonation wave exceeding a certain critical value, then failure of the wave results.

For example, this critical curvature can be

determined in the three-dimensional case—say, for the critical tube diameter situation—and the $d_c = 13\lambda$ correlation can then be explained. Consider a planar detonation wave characterized by a hydrodynamic thickness Δ_H emerging through the tube opening into the unconfined environment. The hydrodynamic thickness Δ_H represents the equivalent thickness of a "one-dimensional" detonation wave where equilibrium conditions prevail at the C-J plane situated at a distance of one hydrodynamic thickness from the leading front. The hydrodynamic thickness, as reported by Vasiliev et al. [11] and Edwards et al. [12], ranges from about 3 to 5 cell lengths in extent. Taking the observed relation between the cell length l and the cell width λ as $l \approx 1.6\lambda$ gives Δ_H equal to about 5-8 cell widths λ . Taking an average value, one can relate Δ_H and λ as $\Delta_H \approx 6.5\lambda$. With regard to transmission, it is plausible to postulate that if the rarefaction waves do not reach the core of the fully exposed wave (i.e., Δ_H thick) by the time the wave has propagated a distance Δ_H , then the detonation wave will survive. In other words, for successful transmission the detonation must have an unattenuated portion at least of the order of one hydrodynamic thick-

ness. This is compatible with the concept advanced by Urtiew and Tarver [13] of an unattenuated core. For the unattenuated detonation core to be of the order of $\Delta_H \cong 6.5\lambda$ (as contrasted with $\lambda/2$, proposed by Urtiew on the basis that the core represents the minimum constant area tube diameter), the detonation front would have to propagate a distance of $2\Delta_H$. In other words, this implies that the wave Δ_H thick having traveled the distance Δ_H is now $2\Delta_H$ beyond the tube exit and that it has traversed this distance by traveling at the C-J velocity D . Therefore, the time of detonation travel τ_D beyond the tube exit can be written as $\tau_D = 2\Delta_H/D$. On the other hand, the time for the radial penetration of the rarefaction waves is $\tau_R \cong d_c/(2c_1)$, where c_1 is the acoustic speed in the product gases. Thus, the criterion for successful transmission is that τ_D must be just less than τ_R , or in the limit $\tau_D = \tau_R$, which leads to

$$\frac{2\Delta_H}{D} = \frac{d_c}{2c_1}$$

For most detonable mixtures $D \cong 2c_1$, hence $d_c \cong 2\Delta_H$, and therefore $d_c = 13\lambda$, which is in perfect agreement with experiment. It follows that the local radius of curvature at the forward tip of the diffracted wave is $R \cong \Delta_H = d_c/2 = 6.5\lambda$ and can be taken as representative of the curvature imposed by a three-dimensional expansion.

Analogous reasoning would be applicable to, say, a square orifice with some equivalent radius of curvature proportional to the average of the minimum and maximum radial dimensions of the square opening,

$$R = \frac{1}{2} \left[\frac{W + \sqrt{2}W}{2} \right] = \frac{d_{eff}}{2} = 6.5\lambda,$$

which immediately yields $W_c/\lambda = 10.8$, which is in accord with the present experimental results and the results of Mitrofanov [2] and Edwards [3].

One can extend the above arguments to the two-dimensional case. In the case of the rectangular orifice of large aspect ratio ($L/W > 7$), the

planar wave diffracting through the opening transforms into a cylindrical wave. For the same critical curvature, by simple geometric considerations, namely,

$$\kappa_{cyl} = \kappa_{sph} = \frac{1}{A} \frac{dA}{dR},$$

one can easily show that the radius of curvature for the equivalent (i.e., same curvature κ) cylindrical wave would be one-half that for the spherical case, so that

$$R_{cyl} = \frac{R_{sph}}{2} = \frac{\Delta_H}{2} = 3.25\lambda = W_c,$$

which again is in agreement with the present results. For the intermediate values of L/W , it is of interest to see whether the correlation $d_{eff} = 13\lambda$ as given by Eq. (3) for the polygon applies as well. For a rectangular slot, the effective diameter is related to the length L and the width W of the slot as follows:

$$d_{eff} = \frac{1}{2} (W + \sqrt{W^2 + L^2}). \quad (6)$$

Replacing d_{eff} by 13λ , the ratio W_c/λ can be obtained:

$$\frac{W_c}{\lambda} = \frac{26}{1 + \sqrt{(L/W)^2 + 1}}. \quad (7)$$

The above expression gives the limit as $L/W \rightarrow 1$ of $W_c/\lambda = 10.8$ as already discussed for the square orifice. For the value of $L/W = 7$, Eq. (7) yields $W_c/\lambda = 3.2$, which is in accord with the experimental result of $L/W = 7$, at which point the approach to the asymptotic value is observed to occur. Predictions from Eq. (7) are shown by the dotted line in Fig. 11 and appear to give the correct trend for the intermediate values of L/W in the approach to the asymptotic limit. Beyond $L/W = 7$, the curve based on Eq. (7) begins to deviate. This is not surprising as now two-dimensionality is being approached and Eqs. (6) and (7) are only valid in the context of the three-dimensional geometry.

4. CONCLUSIONS

The present study of the transmission of a planar detonation from orifices of different geometrical shapes into an unconfined medium has yielded a number of interesting results. The fact that the transmission through an orifice (where the reflected detonation from the orifice plate gives rise to higher stagnation pressures and hence influences the structure of the downstream flow) has the same requirement as that from a straight tube indicates that the reinitiation phenomenon is a very local process, occurring in the immediate vicinity of the detonation front itself. Thus, properties at the wave front (such as the wave curvature) play a more important role than the gas dynamic processes of the product gases in the wake. The results for the transmission through different geometrically shaped orifices lead to the concept of an effective diameter d_{eff} , which is the mean value of the longest and shortest characteristic dimensions of the orifice. The universal correlation based on this effective diameter $d_{eff} \cong 13\lambda$ suggests that the ability of a detonation to evolve without a failure requires a certain minimum limit for the wave front curvature. In other words, as the wave emerges from the tube or orifice and is subjected to lateral expansion, the initially planar front will become curved as the rarefaction waves penetrate into the core of the detonation. Failure occurs when the mean curvature of the attenuating detonation exceeds a certain limit. The mean curvature being proportional to the average linear dimensions of the wave results in the successful correlation of $d_{eff} \cong 13\lambda$. Based on the effective diameter correlation, formulas have been developed to provide the transmission criteria for arbitrarily shaped orifices (i.e., polygonal, elliptical, rectangular), and experimental results agree with these general expressions. Of particular interest in the present study are the results of the two-dimensional limit when one characteristic dimension becomes much larger than the other. Experiments conclusively indicate that the two-dimensional limit gives the shorter characteristic dimension a critical limiting value of about three cell diameters (i.e., $W_c \cong 3\lambda$). This limiting

value becomes independent of the longer dimension when the ratio $L/W \gtrsim 7$. The observed change in the transmission criterion from $W_c/\lambda \cong 10$ for the square orifice to $W_c/\lambda \cong 3$ for a rectangular orifice of large aspect ratio ($L/W > 7$) can be explained in terms of the recently proposed wave curvature model of Lee. Lee's model postulates a minimum critical wave curvature criterion for transmission. Since for the same curvature the associated radius of curvature is less for a cylindrical wave than a spherical wave, such direct geometric considerations clearly confirm the experimentally observed effects.

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gas dynamic flow structure in the wake. Expressions are developed to provide estimates for the critical transmission dimensions for arbitrarily shaped openings. For the two-dimensional limit when one of the characteristic linear dimensions becomes very large compared to the other, it is found that the transmission is based on a critical value for the shorter dimension of the order of 3 times the cell diameter. This result is in accordance with the recent large scale experiments of Benedick in two-dimensional channels of an aspect ratio L/W as large as 35. These observed results can also be successfully explained in terms of the critical wave curvature criterion.

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APPENDIX III

LARGE-SCALE EXPERIMENTS ON THE TRANSMISSION OF FUEL-AIR DETONATIONS
FROM TWO-DIMENSIONAL CHANNELS*

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ABSTRACT

Results of large-scale experiments on the transmission of hydrogen-air and ethylene-air detonations from a two-dimensional channel into a large volume are reported. These data confirm the surprising observations of Liu et al. who reported recent laboratory-scale results which indicate that successful transmission of a detonation emerging from a rectangular orifice with a large aspect ratio $L/W \gtrsim 5$, requires that the minimum width of the channel need be only about three detonation cell diameters, i.e. $W_c \approx 3\lambda$. These results are surprising in view of the previous findings of Soloukhin and Edwards, where for square channels ($L/W = 1$), they observed that $W_c = 10\lambda$.

The apparatus for the large-scale experiments consisted of a two-dimensional confined channel (1.83 m wide x 2.44 m long) with a variable width orifice, to the end of which was attached an inflated polyethylene bag of much larger dimensions to provide an unconfined environment for detonation transmission. Observations were

* Presented at the 9th International Colloquium on Dynamics of Explosion and Reactive Systems, Poitiers, France, July 1983

made with high speed cinematography and with smoked foils to measure the detonation cell size. Our results indicate that a minimum channel width of 3λ is required for successful transmission from channels whose aspect ratios varied from 34 to 8. For channel aspect ratios of less than about 8, the minimum channel width for successful detonation transmission increases continuously from 3λ to 10λ as the aspect ratio approaches 1.

INTRODUCTION

During the past few years, there have been many investigations to determine the critical tube diameter for the transmission of confined planar detonations to unconfined spherical detonations. Both laboratory experiments (Matsui and Lee 1978; Knystautas et al. 1982) as well as large scale field tests have been carried out for a variety of hydrocarbon-air and oxygen enriched mixtures over a range of initial pressures*. The importance of the critical tube diameter is that it is directly related to the dynamic detonation parameters such as critical initiation energy and detonability limits (Lee et al. 1982). Knowledge of the critical tube diameter permits these important dynamic parameters of the explosive to be estimated. The critical tube diameter, d_c , is also found to be directly related to the cell size, λ , of the detonation front via the simple empirical formula, $d_c = 13 \lambda$. This observation was first made by Mitrofanov and Soloukhin (1965) who observed that for low pressure acetylene-oxygen detonations in circular tube, $d_c = 13\lambda$. The significance of this went unnoticed until Edwards et al. (1979) repeated the experiments, confirmed the results and suggested that the law should be valid for other gas mixtures. Subsequently, there have been a number of experimental demonstrations of the validity of this simple empirical law for many different fuel-oxidizer combinations at various initial pressures. Despite the fundamental significance and simplicity of this empirical law, no theoretical justification has appeared. However, extensive

*See Guirao (1982), Moen (1982) and Rinnan (1982).

calculations by Westbrook and Urtiew show a strong correlation between the critical tube diameter (or equivalently the cell size) and the reaction zone thickness as determined by the detailed kinetics of the oxidation process*.

To test the range of validity of this simple empirical law, experiments have been carried out recently by Liu et al. (1983) for detonation transmission through orifices of different geometries. They found that the transmission through an orifice is identical to that through a tube of the same cross-sectional area. For the two-dimensional case of a rectangular orifice of large aspect ratio (i.e., $L/W > 5$), Liu obtained the rather surprising result that the critical channel width W_c for transmission is only about three cell diameters. This result is in disagreement with that reported by Mitrofanov and Soloukhin (1965), as well as Edwards et al. (1979) who gave $W_c \approx 10 \lambda$ for the two-dimensional case. However, both Soloukhin's and Edwards' experiments were based on square channels, whereas Liu's two-dimensional orifice experiments covered a range of aspect ratios $1 < L/W < 15$. For $L/W = 1$, Liu's results agree with Mitrofanov and Soloukhin (1965) and Edwards et al. (1979), $W_c \approx 10 \lambda$.

Due to the fundamental significance of this two-dimensional result reported by Liu, we felt it was desirable to perform large-scale experiments to confirm that $W_c \approx 3 \lambda$. Whereas Liu's experiments were based on rectangular orifice plates of a maximum $L/W \approx 15$, two-dimensional channels with large aspect ratios (e.g., $L/W \approx 30$) can readily be used in field experiments to give a truly

*See Westbrook, (1982), Westbrook and Urtiew (1982), Westbrook and Urtiew (1982), Westbrook (1980).

two-dimensional geometry. The present paper reports the results of a series of such field tests recently carried out at Sandia National Laboratories (New Mexico) to verify the empirical law $W_C \approx 3\lambda$.

EXPERIMENTAL METHOD

The experimental detonation channel consisted of two 25 mm thick steel plates 1.83 m wide and 2.44 m long. The channel width W (the spacing between the plates) was varied by using four spacer blocks placed at each of the four corners of the plates (Fig. 1a). One of the side walls of the channel is covered by a sheet of clear plastic to permit photographic observations of the detonation propagation in the channel, while the opposite side wall of the channel is covered by a plywood sheet. The closed end of the channel was also covered by a sheet of plywood onto which are taped thin strips of sheet explosive for initiating the gas detonation. The quantity of strip explosive ranged from 50 to 140g depending upon the width of the channel. The high explosive strips are centrally initiated by an exploding bridgewire detonator. The plastic and plywood side walls and the initiation end of the channel are blown off during the experiment and have to be replaced for each test. The open end of the channel is connected to a large plastic bag fabricated from 100 μ m thick polyethylene sheets which are taped to the steel plates. The plastic bag was about 3.5 m in length. The filling procedure is to first retract the entire plastic bag into the channel by removing almost all of the air in the system with a vacuum pump; then a premixed hydrogen: air mixture is introduced from a large underground storage tank. The plastic bag is extruded from the channel as the system is being filled

with the explosive gas mixture. The final pressure in the system is slightly above ambient so that the plastic bag remains taut. On windy days or when the hydrogen concentration is rich enough such that the buoyancy of the bag is significant, thin nylon ropes are used to anchor the plastic bag in the horizontal position. The detonation channel is located on a thick concrete pad with the plates extended 0.55 m over the edge of the slab to allow room for the inflated plastic bag. The only diagnostic was high-speed cinematography. Detonation velocity as well as successful transmission or failure can be discerned quite readily from the movie record. Hydrogen-air and ethylene-air mixtures have been used and the desired composition was established in the mixing tank by the method of partial pressures. To measure the detonation cell size, soot was deposited on a thin aluminum sheet by a welding torch burning rich mixtures of MAPP (methyl acetylene, propadiene, propane, butane and propylene) and oxygen. The smoked aluminum sheet was then taped to the bottom steel plate of the channel. We found that the smoked sheet remains in place during the test and the cell size could be determined from it after the top plate of the channel was removed following the detonation.

RESULTS

For the case of hydrogen-air mixtures, the cell size λ as determined from the smoked aluminum sheets placed in the two-dimensional detonation channel is plotted against hydrogen-concentration in Fig. 2. Also shown for comparison are the results for λ obtained at McGill by Knystautas et al. (1982) a circular tube

of 300 mm diameter. For the same hydrogen-air mixture, the present data for λ are generally slightly larger than the corresponding values obtained at McGill (solid line). The slight discrepancy may be accounted for by the difference in the ambient pressures between Albuquerque (~ 1600 m altitude) and Montreal (~ 100 m). Since the cell size is directly proportional to the induction time which is inversely proportional to the initial pressure, the McGill results can be scaled to the lower ambient pressure of Albuquerque, i.e.,

$$\lambda_{\text{Albuquerque}} = \frac{P_{\text{Montreal}}}{P_{\text{Albuquerque}}} \lambda_{\text{Montreal}}$$

Using this correction, there is better agreement between measured cell sizes and the rescaled McGill data (shown as the dotted curve).

Rich mixtures of hydrogen-air were not used in the present series of experiments since the objective was only to verify the empirical law $W_c \approx 3 \lambda$ for two-dimensional channels. Extensive experiments have already been carried out for the rich hydrogen-air mixtures for circular tubes and have been reported previously Guirao (1982). It should be noted that the cell size is indicative of the rate processes in the complex detonation structure and is a very useful parameter. Detonation velocities deduced from the high speed movies were in agreement with the Chapman-Jouguet values within our estimated experimental error.

The normalized critical channel height W_c/λ measured in the present experiments is plotted against the aspect ratio of the channel L/W in Fig. 3. Note that for large aspect ratios, $L/W > 10$, the present results give a value $W_c/\lambda \approx 3$ in approximate agreement with the results obtained by Liu using rectangular

orifices instead of a full two-dimensional channel. Experiments with aspect ratios up to 35 yield the same value for $W_C \approx 3\lambda$. Using different hydrogen concentrations and hence different λ 's and different Chapman-Jouguet states, also gives the same results, $W_C/\lambda \approx 3$. This observation indicates that the critical channel height W_C scales according to the cell size λ , and within the present experimental accuracy does not depend explicitly on the Chapman-Jouguet states. Several experiments were also carried out for 6% ethylene-air mixtures with aspect ratio of 14.5 - 18.5. The result $W_C/\lambda = 3$ was obtained for ethylene as well, reinforcing our conjecture that the scaling law for channels is based only on the cell size λ as has been demonstrated in the case of circular tubes (Knystautas et al. 1982).

In the present two-dimensional geometry, it was also possible to test the symmetry of the transmission process about the centerline of the channel. By retracting the top plate of the channel relative to the bottom plate so that the emergent detonation is supported by one plane, we constructed a half channel (Fig. 1b). By symmetry, transmission should now be possible for a spacing between the plates corresponding to only 1.5λ . Several experiments were conducted using 20 - 20.5% hydrogen-air mixtures with channel widths from 10 to 10.5 cm and a channel length of 1.82 m, yielding $17.5 < L/W < 18.2$. The go - no go results bracketed a critical spacing between the plates of close to 1.5λ , confirming that the two-dimensional transmission mechanism is symmetrical about the channel centerline.

For aspect ratios $L/W < 8$, the present results indicate that the value for W_C/λ rises sharply. A similar trend was also obtained

by Liu for the case of transmission through rectangular orifices, but the increase does not occur until $L/W \lesssim 5$. Why three-dimensional effects begin to become important at larger aspect ratios for the channel than for the orifice plates has not been resolved. Other than this discrepancy, the present channel experiments are in qualitative agreement with the rectangular orifice plate experiments of Liu.

To explain the surprising result that the critical condition for the successful transmission of detonation changes from $W_c = 10\lambda$ for a square channel to $W_c = 3\lambda$ for a rectangular channel of large aspect ratio ($L/W \gtrsim 8$), one can invoke the wave curvature concept proposed recently by Lee et al. (1984). Lee's arguments suggest that when a planar detonation wave propagating in a confining linear tube or channel emerges suddenly into an unconfined geometry, it is subject to a gas dynamic expansion process by virtue of the rarefaction waves that are generated at the edge which then penetrate into the wave front and impose a curvature on the wave. He then defines the critical condition for transmission in terms of the minimum radius of curvature of the diffracted wave, namely; if the rarefaction waves give rise to a curvature of the diffracted detonation exceeding a certain critical value, then failure of the wave results. For a three-dimensional diffraction process the minimum radius of curvature would be of the order of the hydrodynamic thickness Δ_H so that $R = \Delta_H = 6.5\lambda$. Thus for the diffraction of detonation through a square opening, the expansion process is three-dimensional and the curvature imposed on the diffracting wave would be spherical in nature corresponding to some

effective radius of curvature related to the average of the minimum and maximum dimensions of the square opening $R \approx (W + \sqrt{2}W)/2$.

Liu et al. (1983) has shown that for this case $W_c \approx 10\lambda$ in agreement with the experimental results of Soloukhin (1965) and Edwards (1979).

For rectangular channels of high aspect ratio ($L/W > 8$); the planar wave diffracting through the opening transforms into a cylindrical wave. For the same critical curvature, by simple geometric considerations, namely that $\kappa_{cyl} = \kappa_{sph} = (1/A)dA/dR$ one can show that the radius of curvature for the cylindrical wave would be one half that for the spherical case so that $R_{cyl} = R_{sph}/2 = \Delta H/2 = 3.25\lambda$ which is in agreement with the present experimental results.

CONCLUSION

The present series of large scale tests confirm the smaller laboratory experiments of Liu et al. that transmission from confined planar detonations to unconfined cylindrical detonations requires a critical channel height of only about three cell diameters. Furthermore, the channel results are almost identical to the results from a rectangular orifice. This suggests that the re-initiation mechanism is very local and does not depend strongly on the flow structure of the product gas emerging into confined space behind the leading front. The onset of three-dimensional effects at surprisingly large aspect ratios (i.e., $L/W \approx 5$) suggests that quenching by the rarefaction waves may not be a dominant mechanism.

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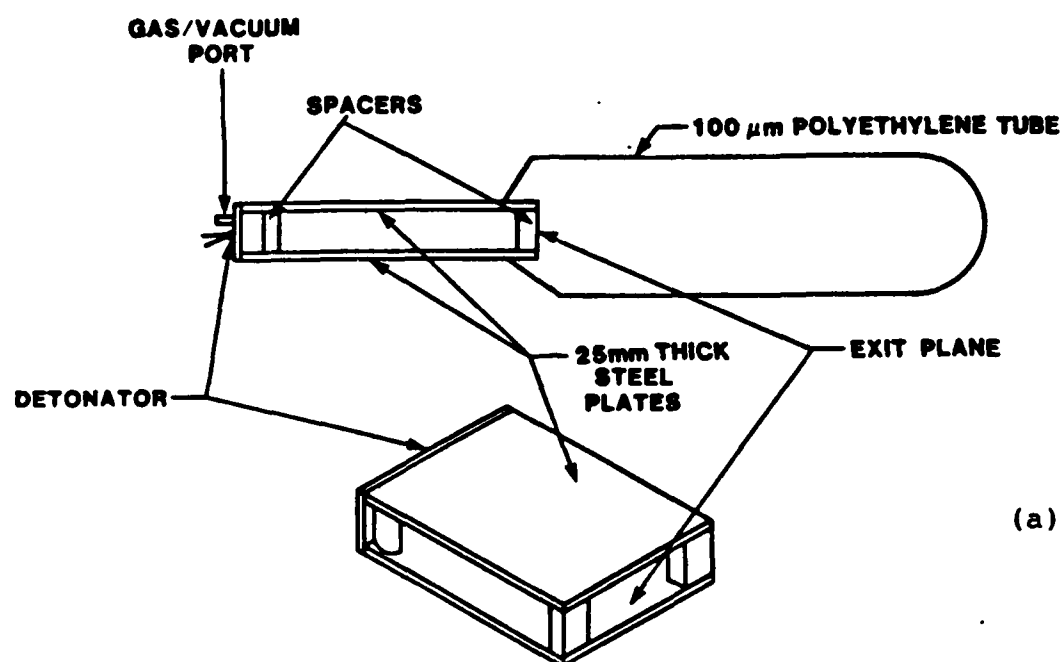
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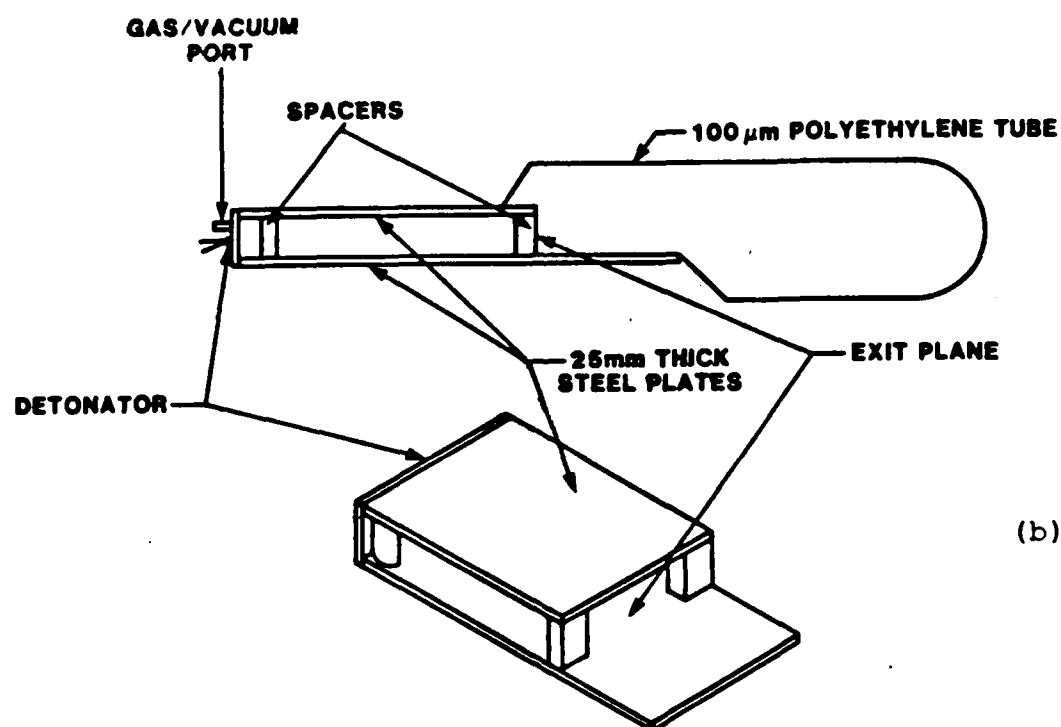
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FIGURE CAPTIONS

- Fig. 1 Two-dimensional channel apparatus with:
 (a) unconfined exit plane
 (b) one degree of confinement at the exit plane.
- Fig. 2 Detonation cell size measured in Montreal,
 normalized for lower ambient pressure in
 Albuquerque, and as measured in channel
 experiments in Albuquerque, all as a function
 of H_2 :Air mixture.
- Fig. 3 Critical channel width normalized to detonation
 cell size, W_c/λ , as a function of the channel as-
 pect ratio, L/W , for detonations emerging from the
 exit plane of the channel into an unconfined
 volume. The data point $3\lambda/2$ is for detonation
 supported by one plane after exiting the channel.



(a)



(b)

Figure 1

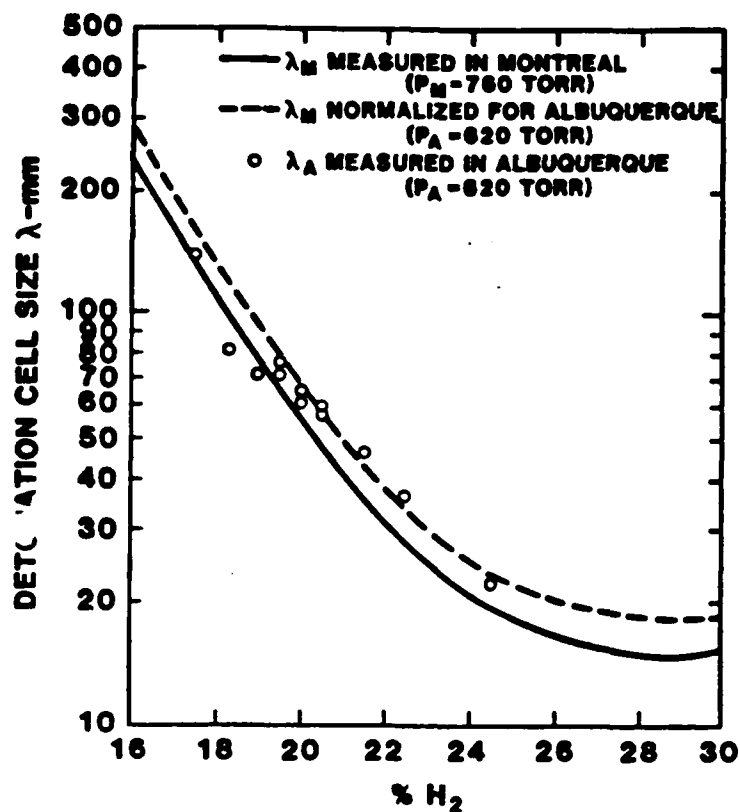


Figure 2

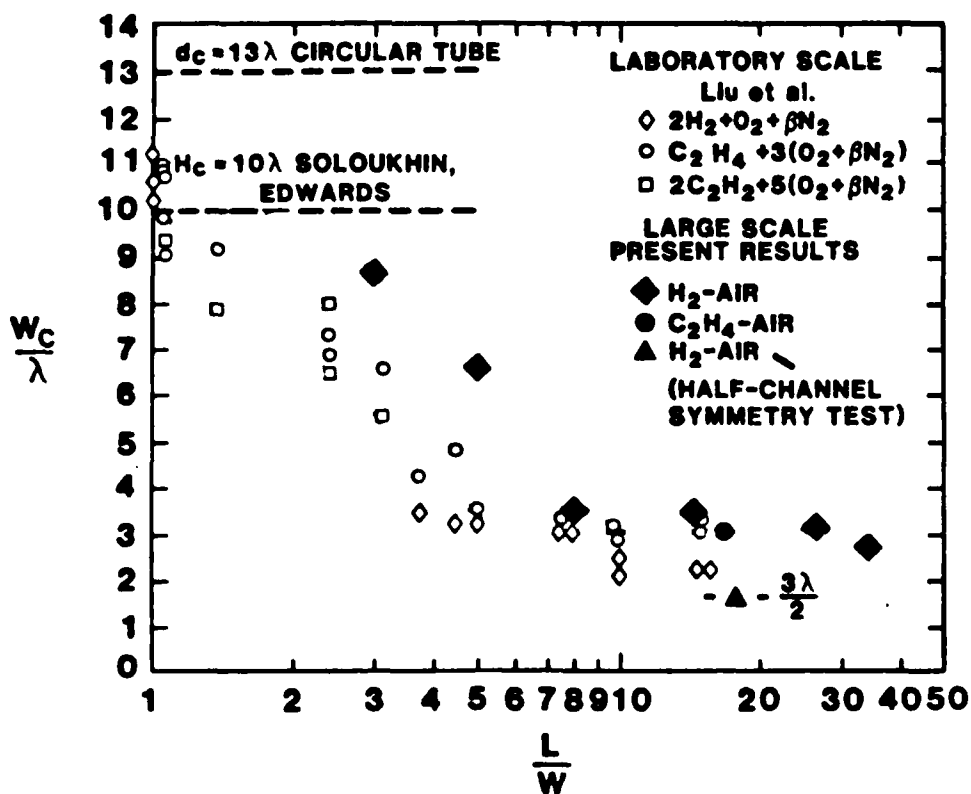


Figure 3

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Recent laboratory scale experiments on the transmission of detonation waves through two-dimensional rectangular orifices have indicated the surprising result that for large aspect ratios ($L/W \approx 10$), the transmission criterion requires a minimum channel width $W_c \approx 3\lambda$, where λ is the characteristic detonation cell size. This appears to be contrary to previous findings by Soloukhin and Edwards where for square channels they had observed that $W_c \approx 10\lambda$. The present paper reports the results of large field scale experiments to confirm this finding and resolve the discrepancy with pre-		

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vious observations as well as to explore the range of its validity. The experiments were carried out in H_2 -air and C_2H_4 -air mixtures for a range of compositions at the New Mexico test site of Sandia National Laboratory. The apparatus consisted of a two-dimensional confined channel (2.36 m wide x 3.15 m long) with a variable width W to the end of which was attached an inflated polyethylene bag of much larger dimension to provide the unconfined environment for the detonation transmission from the confined channel. Observations were made using high speed cinematography and also with the smoked foil technique for detonation cell size measurements. The results indicate that indeed for $5 \lesssim L/W < 34$, $W_c \approx 3 \lambda$ but that for $5 \lesssim L/W < 1$, the required number of cells for the transmission process increases and approaches $W_c \approx 10 \lambda$ for $L/W \approx 1$. Several tests were also carried out to indicate that the transmission phenomenon is symmetrical about the central plane.

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APPENDIX IV

DIFFRACTION OF DETONATION FROM TUBES INTO A LARGE FUEL-AIR EXPLOSIVE CLOUD

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This paper reports on a series of field tests performed at the Canadian Defence Research Establishment Suffield and at Raufoss, Norway, to obtain the critical tube diameter, d_c , for ethylene-air mixtures by investigating the diffraction of detonations from tubes into large plastic bags simulating an unconfined fuel-air cloud. The critical ethylene-air compositions for successful re-establishment of detonation upon emerging from tubes of diameters 0.31 m, 0.45 m, 0.89 m and 1.36 m were determined by monitoring the diffracted detonation wave in the bag. High-speed cinematography of the diffracted wave shows that re-establishment of detonation is via one or more re-ignition centres at sites along the head of the expansion wave which originates at the area change. The characteristic transverse wave spacings, S , associated with the detonations were measured from smoked foils mounted in the tubes. These measurements demonstrate that the empirical relation $d_c = 13 S$ provides a good correlation between the critical tube diameter and the cell size over a wide range of ethylene-air compositions. However, a better understanding of the coupled chemical-gas-dynamic processes within the cell is required in order to clarify the link between cell size and chemical kinetics.

In addition to the open tube tests, transmission of detonations through circular orifice holes was investigated in both ethylene-air and acetylene-air mixtures. Based on these tests it was concluded that the critical tube and critical orifice diameters are equal.

1. Introduction

Two key properties which characterize the detonability of an explosive mixture are the critical energy, E_c , required to initiate detonation in the mixture and the critical tube diameter, d_c , required for a planar detonation emerging from a tube to transmit to an unconfined spherical detonation. Matsui and Lee¹ have proposed that either E_c or d_c can be used to assess the relative sensitivity of explosive mixtures to detonation. In fact, they propose a simple relation between these two parameters based on a work-done model. On a more fundamental level, these properties are related to the detailed coupling between chemical energy release and gasdynamics which is also responsible for the three-dimensional transverse wave or cellular structure of detonation waves.

The mechanism by which the transverse waves are excited and maintained is not completely understood. However, the recent detailed observations on the gasdynamics and chemistry within a detonation cell by Libouton et al.² have provided new insight into the re-initiation process at the end of the cell. It is this re-initiation which ensures the maintenance of the cell structure characteristic of a self-sustained detonation. The delay to re-initiation and thus the length of a detonation cell is controlled by the chemical kinetic properties of the explosive mixture. The critical tube diameter, on the other hand, is a measure of minimum detonation size required for the regeneration or excitation of transverse waves as the planar detonation wave emerging from the tube is being quenched by the transversely advancing expansion waves.

The first observation that a minimum tube di-

ameter is required for an established detonation to emerge from a tube and become a spherical detonation in an unconfined cloud was made by Zel'dovich et al.³ Mitrofanov and Soloukhin⁴ later observed that the critical diameter for oxy-acetylene systems is 10 to 13 transverse wave spacings. Edwards et al.^{5,6} have provided further evidence that a minimum number of transverse waves is required for successful transmission. They also suggest that the number of transverse waves required depends only on the geometry of the tube (i.e., $d_c = 13 S$ for a circular tube and $d_c = 10 S$ for a planar channel and not on the detonative system. The relation $d_c = 13 S$ for critical transmission from circular tubes has recently been confirmed for a variety of oxygen-enriched fuel-air mixtures by Knystautas et al.⁷ However, the direct tests of this empirical relation have been limited to mixtures having relatively small cell structure ($S \leq 16$ mm). Edwards et al.⁶ present some evidence that detonations in systems with large and less regular cell structure require more transverse waves to ensure transmission of the diffracted wave. It is therefore important to examine the relation between cell size and critical tube diameter for larger diameter tubes.

In the present tests, the critical C_2H_4 -air compositions for successful re-establishment of detonation upon emerging from tubes of diameters 0.31 m, 0.45 m, 0.89 m and 1.36 m were determined by monitoring the diffracted wave in the bag using ionization-gap probes and pressure transducers. In addition, high-speed cinematographic records of the

diffracting detonation wave were obtained using two high-speed cameras. The detonation cell structure was monitored by the smoked-foil technique.

2. Experimental Details

The experimental field facility at DRES has been described in detail elsewhere.^{8,9} The facility is centred around an 18.3 m \times 7.6 m concrete test pad onto which the experimental apparatus can be mounted. A photograph of the test pad with a typical test section used in the critical tube diameter tests is shown in Fig. 1. The test sections for these tests consisted of steel tubes connected to large plastic bags constructed from 0.13 mm thick, seamless, extruded polyethylene tubing. The dimensions of tube-bag configurations used in the present tests are also shown in Fig. 1.

At DRES, the test gas (CP grade, 99.5% pure ethylene) was mixed with the initial air in the test volume by a multipath re-circulation system using a high-capacity centrifugal blower. The composition and mixture homogeneity in the test volume were verified by continuously analyzing samples taken from four ports positioned strategically along the apparatus using a calibrated "Wilks Miran 80" infrared gas analyzer. This sampling system is adequate to guarantee the concentration and homogeneity of the mixture to within $\pm 0.05\%$ C_2H_4 . Once the desired fuel-air composition had been attained, detonation was initiated by using a De-

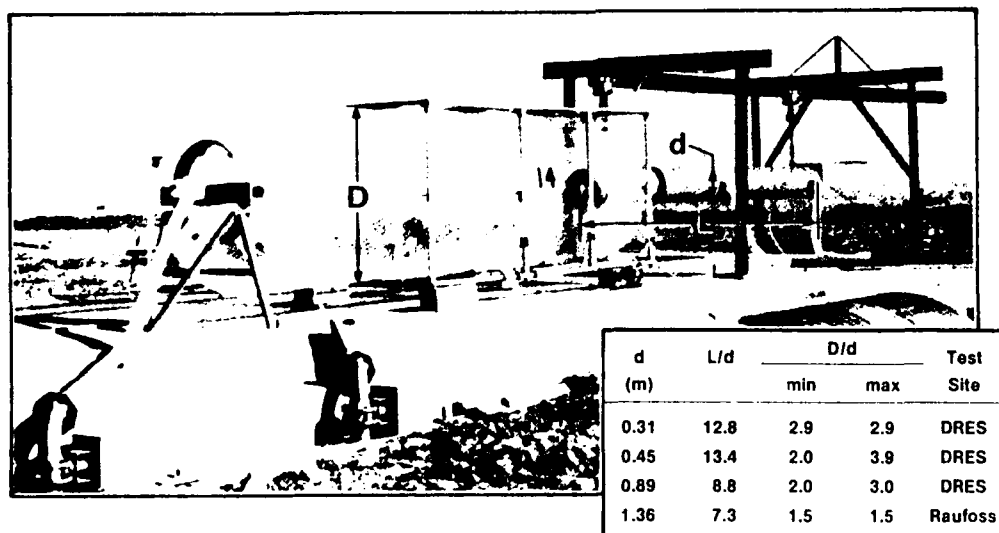


FIG. 1. Photograph showing the DRES testing facility with a detonation tube (0.45 m diameter) and polyethylene bag (1.75 m diameter) test section. Dimensions for each of the four critical tube configurations investigated appear in the table.

tasheet initiator charge mounted on the plywood sheet which covered the ignition end of the tube.

Four types of diagnostics were employed in the DRES tests. Eight piezo-electric pressure transducers (PCB 113A24) were positioned along the apparatus. Pressure-time signatures and velocities obtained from the pressure records were used to determine success or failure of transmission from the tube to the bag. In addition, a 20-channel ionization-gap probe system, together with a time-of-arrival counter, was used to independently measure wave velocities. Cinematographic records were obtained by using a "Hycam" camera (15,000–18,000 half frames per second) looking normal to the direction of propagation and a "Fastax" camera (~5,000 full frames per second) looking at a 45° angle to the direction of propagation. Finally, 350 mm × 500 mm smoked foils mounted in the tube near the exit to the bag were used to record the detonation wave structure. Each foil was made by applying a thin layer of carbon soot to a polished tin sheet.

The experimental configuration, diagnostics and procedure used in the Raufoss tests were similar. In addition to open tube tests, transmission through circular orifice holes was investigated. Each orifice hole was cut in a 20 mm thick plywood disc mounted over the exit end of the tube. These transmission tests were performed using both ethylene-air (C_2H_4 -air) and acetylene-air (C_2H_2 -air) mixtures.

The gas flow system used at Raufoss was based on the flow displacement method. Five test volumes were displaced to guarantee the desired mixture composition. Infrared analysis was employed as in the DRES tests. However, only the input gas mixture was sampled. The estimated uncertainty in mixture composition in the test section was $\pm 0.1\%$ fuel. Pressure transducers, ionization-gap probes and high-speed cinematography were the principal diagnostics. Smoked foils were not used.

3. Results and Discussion

(i) Critical Tube Diameter and Transverse Waves

Selected frames from a high-speed (~15,000 frames/sec) cinematographic record showing the successful transmission of detonation from a 0.89 m diameter tube into a large 1.75 m diameter bag simulating an unconfined gas cloud are shown in Fig. 2. Frame 1 shows the planar detonation wave just as it emerges from the tube. Frame 2 shows the shrinking planar detonation core as the expansion waves which originate at the tube exit sweep in towards the centre quenching the detonation in their wake. The head of the expansion fan reaches the centre at a distance of approximately one tube diameter from the tube exit. Unless re-ignition, capable of

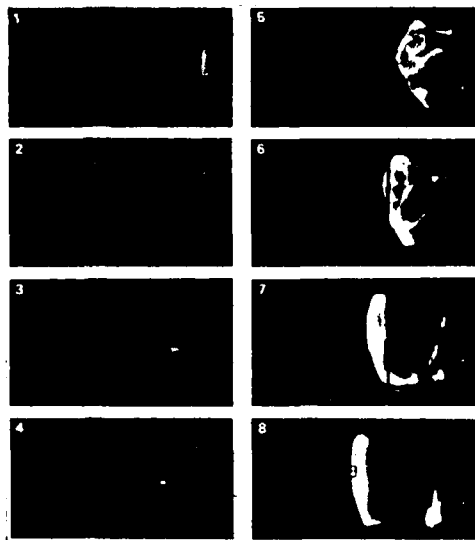


FIG. 2. Selected frames from a high-speed cinematographic record (~15,000 frames/sec) showing successful transmission of detonation from a confined tube of 0.89 m diameter to a large gas bag of 1.75 m diameter simulating an unconfined environment (0.2 msec between frames).

re-establishing the detonation in the adverse expansion, occurs somewhere behind the leading shock wave prior to this, the detonation fails to re-establish itself and the explosive mixture in the bag is consumed by a deflagration. For the test shown in Fig. 2, re-ignition is seen to occur in Frame 3 at an ignition nucleus in the centre of the bag. The subsequent formation of detonation bubbles which sweep across the entire cross section of the bag are clearly seen in Frames 4–6 of the sequence. The final two frames of the sequence show a fully established detonation in the bag.

Fig. 2 represents a typical sequence showing close to critical re-establishment of detonation after diffraction from a tube. If the sensitivity of the mixture is reduced for the same diameter tube, no re-ignition centres are observed and the detonation fails to re-establish itself in the bag, while with more sensitive mixtures re-ignition occurs closer to the tube exit, usually at more than one ignition centre at the edge of the shrinking detonation core. Only for mixtures much more sensitive than critical composition does the detonation wave transmit directly from a planar wave to a hemispherical wave without the appearance of re-ignition centres.

The above observations are in agreement with the laboratory Schlieren and smoked-foil observations on the diffraction of detonations in low pressure oxy-acetylene reported by Edwards et al.^{5,6}

Their records also show that the re-establishment of detonation after an abrupt area expansion is via the formation of re-ignition centres at sites along the head of the expansion which originates at the area change. In spite of the apparent randomness in the appearance of these re-ignition centres, the critical mixture composition for re-establishment of detonation for a given tube diameter is well defined provided; i) the detonation emerging from the tube is a planar detonation wave no longer influenced by the blast wave from the initiating charge, and ii) the plastic bag is sufficiently large so that it does not influence the transmission process.

In the present tests, care was taken to ensure that a steady-state planar detonation had been established and that the initiating charge did not influence the critical fuel-air composition. This was accomplished by varying the initiation energy once the critical composition had been identified and by carefully monitoring both the pressure and velocity of the detonation wave in the tube.

In order to ensure that wave interactions with the plastic bag did not influence the transmission, tests were done using plastic bags of different sizes. The re-establishment of detonation due to wave interactions with the plastic bag was observed on the high-speed film records from only one test (0.45 m tube into a 0.89 m plastic bag at 6.3% C_2H_4). The apparent Go outcome of this test has therefore been discounted.

The results from the critical tube diameter tests are summarized in Fig. 3, where the Go - No-Go outcome of tests with different diameter tubes is shown as a function of % C_2H_4 by volume. Also shown in this figure are the results from the 1980 Raufoss tests.¹⁰ Except for the 0.45 m tube, the results from the two test series are in good agreement considering the different experimental procedures and conditions at DRES and Raufoss. The Raufoss results indicate successful re-establishment of detonation in leaner mixtures for a given tube diameter. This could be due to the fact that air containing impurities in the form of oil mist from an air compressor was used in the Raufoss tests. These impurities could have an effect on the mixture sensitivity and also on the monitoring of fuel concentration. At DRES, atmospheric air was used in all the tests.

It is important to note that no detonation re-establishment was observed with a 0.31 m tube up to an ethylene concentration of 10.5%. Since it is very unlikely that re-establishment will occur in even richer mixtures, we can conclude that the critical tube diameter is larger than 0.31 m for any C_2H_4 -air mixture.

The solid and chain-dotted curves shown in Fig. 3 are theoretical predictions based on a correlation proposed by Moen et al.¹¹ for initial pressures of 101.3 kPa and 92.5 kPa, respectively; the latter

pressure being the mean atmospheric pressure on the DRES range where most of the tests were done. The dashed curve is a similar theoretical prediction using a more detailed chemical reaction scheme proposed by Westbrook.^{12,13}

All of these predictions are based on empirical relations between the critical tube diameter, d_c , the transverse wave spacing, S , and the induction zone length, Δ , calculated for the hypothetical one-dimensional C-J detonation. It is assumed that the detonation cell size is determined by the chemical induction time, τ , behind a shock wave propagating at the C-J velocity, with S being directly proportional to the induction-zone length $\Delta = \tau u$, where u is the post-shock relative particle velocity. When combined with the empirical 13 S formula for d_c , this gives $d_c = 13A\tau u$, where A is the proportionality constant relating S and Δ . Thus, once the C-J shock parameters and the corresponding induction times are known as functions of mixture composition, the critical tube diameters are determined by normalizing at one composition. For the ethylene system Moen et al.¹¹ use the induction-time formula of Hidaka et al.¹⁴

$$\log_{10} \tau [O_2] = -11.45 + 27.5 \times 10^3 / 4.58T,$$

where τ is the induction time in seconds and $[O_2]$ is the oxygen concentration in moles/liter. They then obtain $A = 480$ by monitoring the onset of near-limit phenomena in circular tubes and find that the predicted critical tube diameters for C_2H_4 - O_2 - N_2 mixtures having different nitrogen to oxygen ratios β ($0 \leq \beta \leq 3$) are in good agreement with experimental results. As can be seen in Fig. 3, the predicted d_c for C_2H_4 -air mixtures, using the same value for the constant of proportionality, are also in reasonable agreement with experimental results over a fairly wide range of C_2H_4 concentrations. However, the predicted d_c based on Westbrook's model is out by more than a factor of two at 4.5% C_2H_4 when normalized to $d_c = 0.36$ m at stoichiometric composition (6.54% C_2H_4). Based on C_2H_4 - O_2 - N_2 laboratory results¹¹ Westbrook's model predicts a d_c larger than 0.36 m at stoichiometric composition, so that agreement with all available experimental data on the ethylene system is not improved by re-normalizing to obtain better agreement with the present lean C_2H_4 -air results.

There are two reasons for the difference between these two model predictions. Westbrook's results are obtained using a detailed chemical reaction scheme for ethylene oxidation which appears to predict an effective activation energy much larger (~48 kcal) than the 27.5 kcal obtained by Hidaka et al.¹⁴ from shock tube studies on C_2H_4 - O_2 mixtures highly diluted by argon. Furthermore, Westbrook uses the equilibrium temperature behind the shock wave to calculate the induction time. This

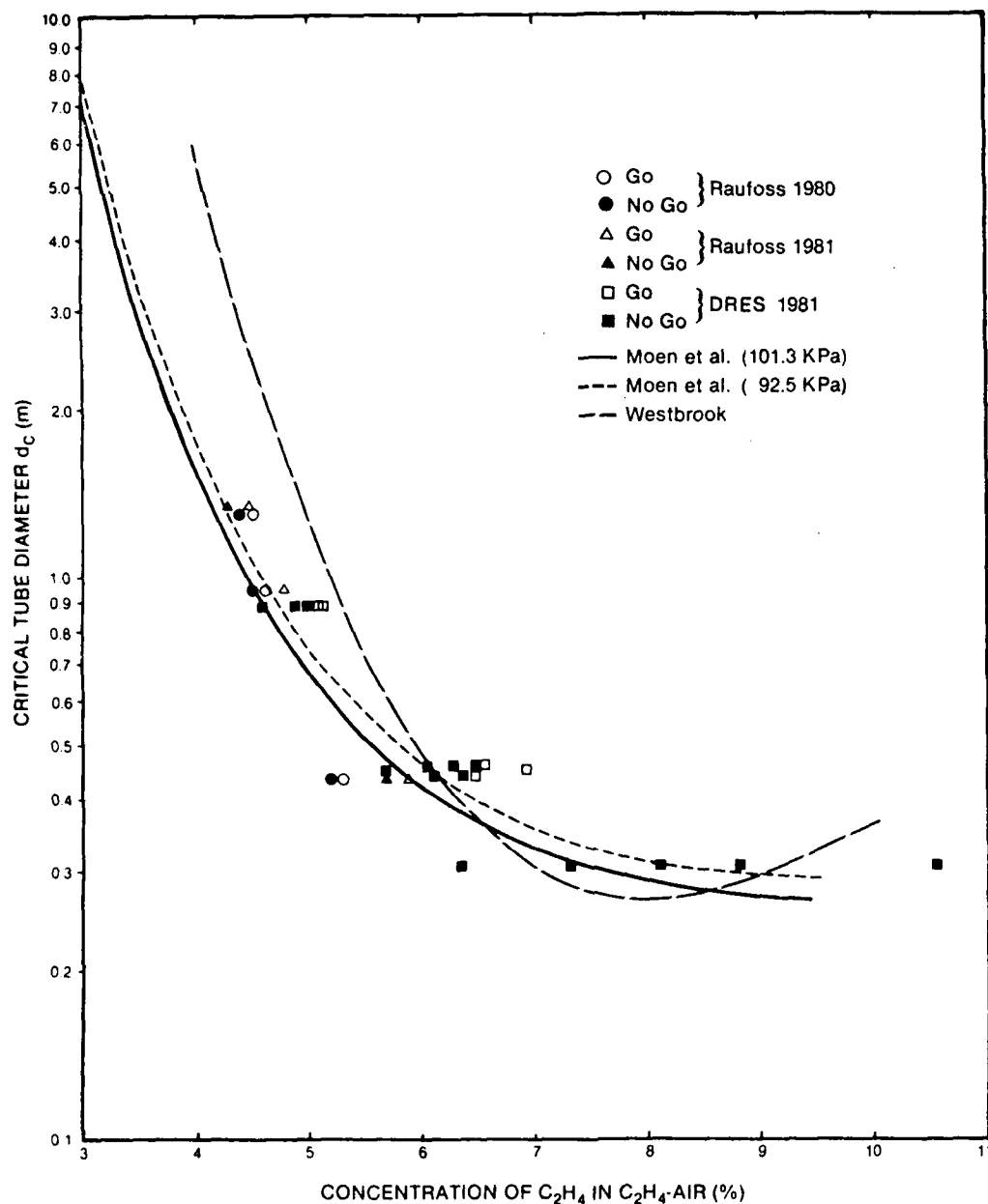


FIG. 3. Critical tube diameter versus concentration (%C₂H₄ in C₂H₄-air by volume). Raufoss 1980—Ref. 10; Moen et al.—Ref. 11; Westbrook—Refs. 12, 13.

temperature is considerably lower than that calculated assuming constant specific heat ratio across the shock as Moen et al. have done.

Westbrook's correlations of critical tube diameters, and also of critical initiation energies using the Zeldovich³ relation $E_c \propto \Delta^3$, for a variety of

fuel-air mixtures are quite impressive.^{12,13} However, the tests of these correlations have been limited to relatively sensitive mixtures with $d_c \leq 0.2$ m and $E_c \leq 0.1$ kg of tetryl. The present results, as well as the 1980 initiation energy results,¹⁵ show that the correlations are not in good agreement

with experiment over a wider range of mixture sensitivity. In view of the rather arbitrary choice of C-J conditions for determining the cell size, this is not surprising. It is well established that the shock velocity within a detonation cell varies from an initial value of about $1.6 U_{CJ}$ to a minimum of about $0.8 U_{CJ}$ near the end of the cell. Although C-J conditions represent the average conditions in the cell, the re-initiation process within the cell observed by Libouton et al.² does not occur at these average conditions. A better understanding of the coupled chemical gasdynamic processes within the cell is clearly required in order to properly model the link between cell size and chemical kinetics.

The link between cell size and critical tube diameter is also not understood. However, in this case the 13 S relation continues to hold for less sensitive mixtures. The transverse wave spacings measured in the tubes are plotted as a function of $\%C_2H_4$ by volume in Fig. 4. The results are in good agreement with the measurements of Bull et al.,¹⁶ Borisov¹⁷ and Knystautas et al.⁷ at stoichiometric composition, and also with the results of Moen et al.¹¹ obtained by monitoring the onset of near-limit phenomena in tubes. The curve in this figure corresponds to the solid curve in Fig. 3 with

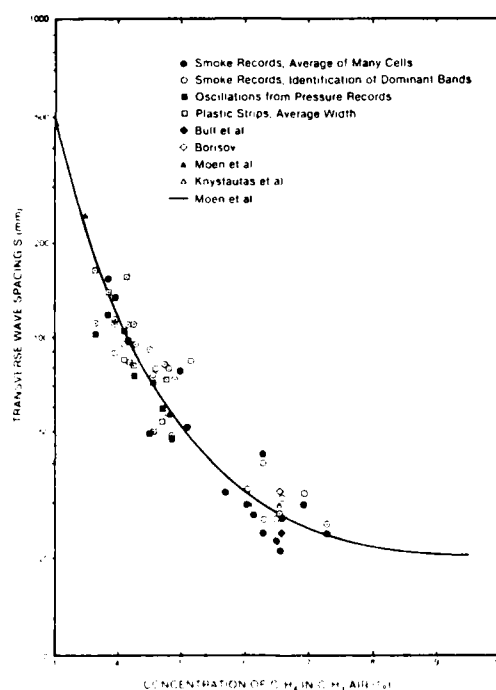


FIG. 4. Transverse wave spacing versus concentration ($\%C_2H_4$ in C_2H_4 -air by volume). Bull et al.—Ref. 16; Borisov—Ref. 17; Moen et al.—Ref. 11; Knystautas et al.—Ref. 7.

$S = d_c/13$. Although there is considerable scatter in the data, the empirical 13 S formula provides a reasonably good correlation over a wide range of C_2H_4 -air compositions. The scatter in the data is partly a reflection of the complex structure observed on many of the smoked-foil records. The structure typically consists of large cells superimposed on a smaller much more irregular structure, so that the identification and measurement of the relevant cell structure can be difficult.

A typical smoked-foil record, together with a sketch illustrating our interpretation of the record, is shown in Fig. 5. Two independent measuring techniques were used to determine the most representative transverse wave spacings from these records. The first method was simply to take the average value of the cell width from a large number of cells. In this approach some judgement as to what constitutes a cell is required. This is avoided to some extent in the second method which is based on identification of dominant diagonal bands of similarly shaped cells. For low mode number detonations in the tube the characteristic cell size can also be deduced from the frequency of the pressure oscillations behind the leading detonation wave.

An interesting observation is that the width of the longitudinal plastic strips produced by a detonation in the bag appears to be equal to the transverse wave spacing. The widths of the plastic strips observed for different C_2H_4 -air compositions are included in Fig. 4. Many of the plastic strip results were obtained in a subsequent test series involving the transmission from a tube into a plastic bag of the same diameter.¹⁸

ii) Transmission Through Orifices

As long as the re-establishment of detonation upon diffraction is localized (i.e., confined to a region in the immediate neighbourhood of the diffracted detonation core where the complex shock reflections from an orifice plate would not influence the gasdynamic flow field), the critical conditions for transmission through a circular orifice should be the same as those for a circular tube. The equality of the critical tube diameter and critical circular orifice diameter was first demonstrated in a series of laboratory tests with oxygen-enriched ethylene-air and acetylene-air mixtures for orifices of diameters 89, 51, 25 and 13 mm at the end of a 200 mm diameter tube using the apparatus described by Knystautas et al.⁷ The large-scale tests with orifices of diameters 0.96 m, 0.44 m and 0.20 m at the end of the 1.36 m diameter tube provide evidence that this equality also holds for less sensitive fuel-air mixtures. The Raufoss 1981 experimental points at $d_c = 0.96$ m and 0.44 m in Fig. 3 were obtained from these transmission tests. In view of the significant disagreement between the

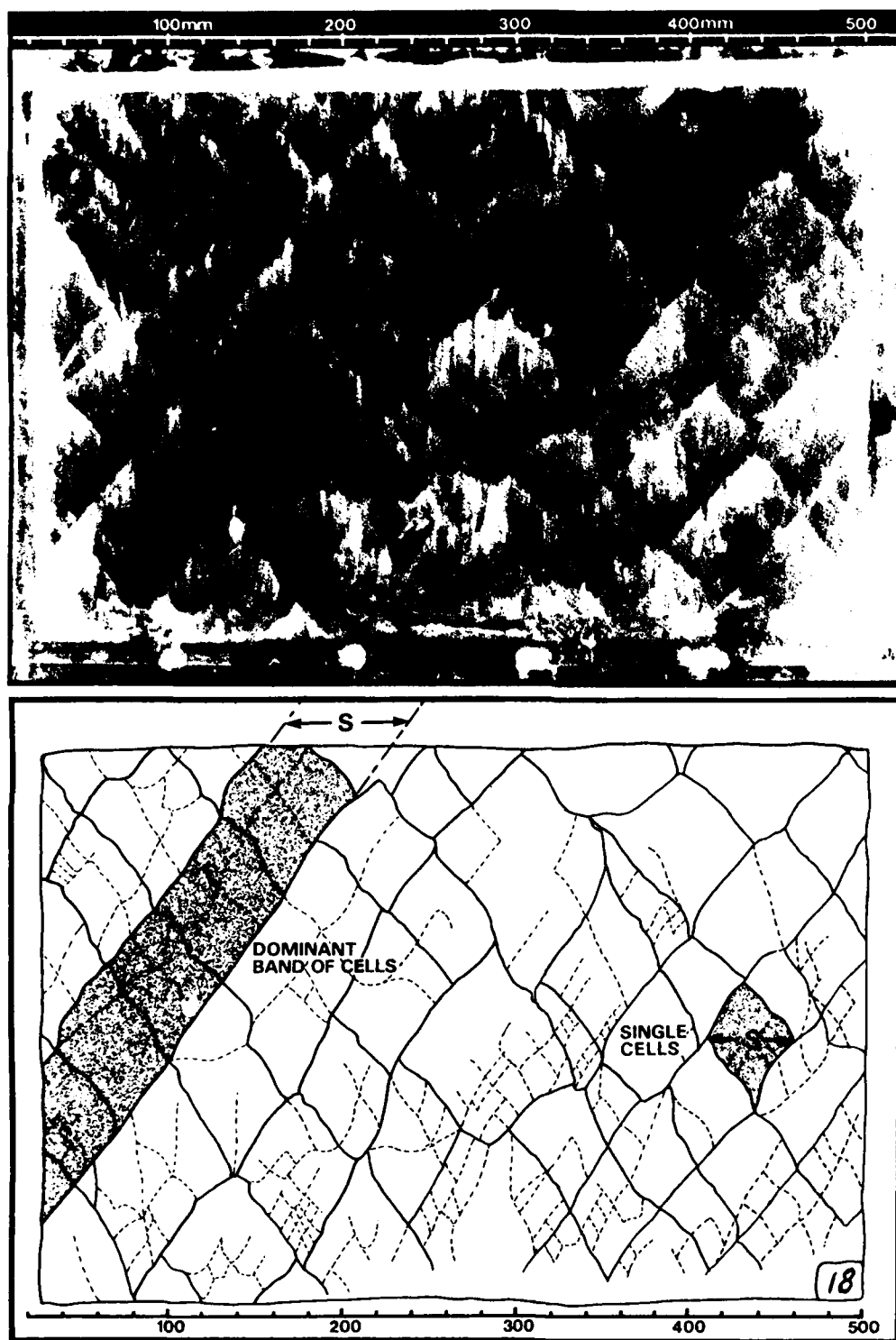


FIG. 5. Typical smoked foil (4.6% C_2H_4 in a tube of 0.89 m diameter) with interpretation sketch illustrating both a single cell and a dominant diagonal band of cells.

present results and the 1980 Raufoss results¹⁰ at $d_c = 0.45$ m and the fact that only a Go result is available for the 0.96 m diameter orifice, these results are not very convincing evidence of the equality. More convincing evidence was obtained for C_2H_2 -air mixtures with orifice holes of diameters 0.44 m and 0.20 m. As can be seen in Fig. 6, these results agree with the 1980 critical tube results¹⁰ within the experimental uncertainty in fuel concentration ($\pm 0.1\%$ C_2H_2). Also shown in Fig. 6 are critical tube results at stoichiometric composition (7.75% C_2H_2) obtained by Knystautas et al.,⁷ and by Freiwald and Koch,¹⁹ together with the prediction based on 13 S from Bull et al.¹⁶

The theoretical curves shown in Fig. 6 were derived in the same manner as those for C_2H_4 -air mixtures in Fig. 3. The solid curve was arrived at by the procedure of Moen et al.¹¹ using the induction-time formula of White²⁰ with an activation energy of 17.3 kcal, and the dashed curve was obtained using Westbrook's model for C_2H_2 oxidation.^{12,13} As was the case for ethylene-air mixtures, Westbrook's model appears to predict too strong a variation of d_c with fuel concentration.

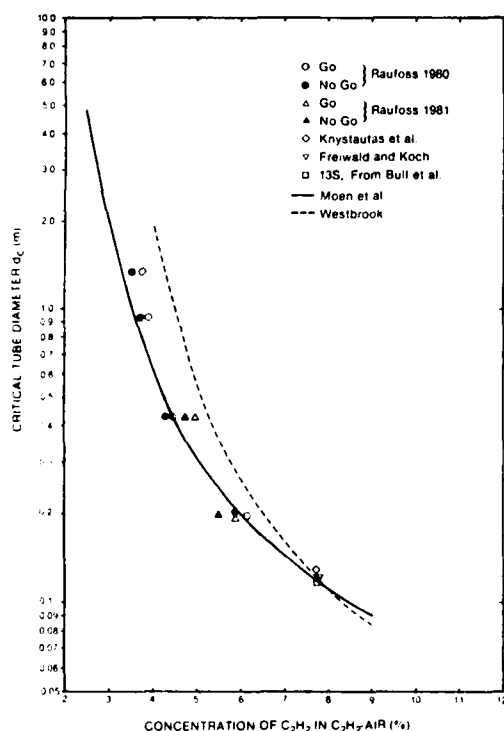


FIG. 6. Critical tube diameter versus concentration (% C_2H_2 in C_2H_2 -air by volume). Raufoss 1980—Ref. 10; Knystautas et al.—Ref. 7; Freiwald and Koch—Ref. 19; Bull et al.—Ref. 16; Moen et al.—Ref. 11; Westbrook—Refs. 12, 13.

4. Conclusion

Large-scale field tests on the diffraction of detonations in ethylene-air mixtures have provided key data on the detonability of ethylene-air for a range of fuel concentrations from 3.6% to 10.5% C_2H_4 by volume. These tests show that the critical tube diameter for any ethylene-air mixture is larger than 0.31 m. The critical tube diameter near stoichiometric composition (6.54% C_2H_4) is 0.45 m, which is slightly larger than the values of 0.36 m to 0.38 m obtained by extrapolating from laboratory results in oxygen-enriched ethylene-air mixtures.^{7,11} This difference may be partly due to the low atmospheric pressure (92.5 kPa) on the DRES range where most of the large-scale tests were performed. In the largest 1.36 m diameter tube the critical fuel concentration is found to be 4.5% C_2H_4 . According to the work-done model of Matsui and Lee,¹ the critical initiation energy E_c varies as d_c^3 . Based on this assumption, the present results predict that E_c at 4.5% C_2H_4 is about 30 times that at stoichiometric composition. This prediction is in good agreement with large-scale measurements of the critical energies¹⁵ and thus provides further evidence that either E_c or d_c can be used to determine the relative detonation hazards of explosive mixtures, as originally proposed by Matsui and Lee.¹

Measurements of transverse wave spacings or detonation cell sizes show that the empirical relation $d_c = 13$ S, first discovered for oxy-acetylene⁴ and found to be valid for a variety of oxygen-enriched fuel-air mixtures,⁷ is also valid for fuel-air mixtures with characteristic transverse wave spacings up to about 100 mm. However, the correlation between cell size and chemical kinetics, based on a linear relationship between the transverse wave spacing and the induction-zone length calculated at C-J conditions using the C_2H_4 oxidation scheme of Westbrook,^{12,13} is not in good agreement with the present data. The data are in better agreement with a similar empirical correlation proposed by Moen et al.¹¹ which uses a smaller effective activation energy and a larger temperature to calculate the induction-zone length.

High-speed cinematographic records of the detonation diffraction process show that the re-establishment of detonation, after the abrupt area expansion from a tube to a large bag, is via the formation of localized re-ignition centres at sites along the head of the expansion which originates at the area change. At criticality, re-ignition occurs at a localized ignition centre near the tube axis, approximately one tube diameter from the tube exit. The localized nature of this re-ignition, immediately behind the shrinking detonation core, suggests that the critical conditions for re-establishment of detonation depend only on the frontal area of the emerging planar detonation wave. The equality of the critical tube diameter and the crit-

ical circular orifice hole diameter is further evidence for this conclusion.

Further refinements in both theory and experiment are necessary in order to obtain a more fundamental understanding of the detonability properties of fuel-air mixtures. The present results, together with the critical energy results from previous large-scale tests¹⁵ provide key data for fuel-air mixtures with characteristic detonation length scales (i.e., S and d_c) that cannot be obtained in the laboratory. These data are not only of practical interest for accidental explosions and military applications, but also provide critical tests of theoretical models and correlations linking the detonability properties to the chemical kinetics of the explosive mixture. Based on detailed observations of the detonation diffraction process, the monitoring of detonation structure and comparison of results with theoretical models, we can conclude that a more fundamental understanding of the detonability properties can only be achieved through further elucidation on the nature of the re-ignition process, both in the diffracted detonation wave and at the end of a detonation cell.

5. Acknowledgments

We gratefully acknowledge the assistance of Mr. John Funk and Mr. Stephen Ward, without whose help the tests at DRES could not have been done. We would also like to thank the personnel of the Field Operations Section, the Electronic Design and Instrumentation Group and the Photo Group at DRES for their assistance during the DRES field tests. The valuable assistance of Norsk Hydro in taking care of the gas supply and gas concentration monitoring system during the Raufoss tests is also gratefully acknowledged.

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COMMENTS

C. Westbrook, Lawrence Livermore Lab., USA. This paper is a very fine collection and presentation of experimental data which are of unusual value in developing new models for detonation properties and understanding such phenomena.

However, the use of simplified, particularly one-step, kinetics expressions for induction times in detonations cannot really be justified any longer for fuels where detailed models exist, such as C_2H_4 and C_2H_2 . The Hidaka expression used here was derived for highly dilute mixtures in shock tubes and covered only a very limited range of fuel-oxygen ratio. Its agreement with these experimental detonation data can only be fortuitous.

Both cell size and critical tube diameter for C_2H_4 -air must increase for fuel concentrations in excess of 7-8% C_2H_4 as predicted by my own kinetics model as referenced in this paper. However, using the simplified expression, the curve of d_c vs fuel concentration (Fig. 3) continues to fall even for rich mixtures. The same type of behavior should also occur for C_2H_2 -air, also incorrectly predicted by the simplified mechanism.

Author's Reply. It is certainly true that the justification for using a one-step chemical kinetic model is very weak. However, there is also little justification for using a more detailed chemical model without taking into account the complex 3-D structure of the detonation wave. This is not to say that such modelling is not valuable, but such parameters as the cell size, critical tube diameter and critical energy can only be calculated when a combined gas dynamic-chemical kinetics model, which takes into account the cellular detonation structure,

has been developed. Until such time, the correlation of these parameters using chemical kinetic models is purely empirical, so that the justification for using one model over another can only be determined from experiment. In the particular case of C_2H_4 -air mixtures, the one-step kinetic model appears to correlate the results for lean mixtures. However, as Dr. Westbrook states, it fails to account for the increase in cell sizes for rich mixtures recently observed by Knystautas et al. at McGill University. For C_2H_2 -air, the available data is not extensive enough to determine which correlation is better.

Although, the success of these simplified models, including Dr. Westbrook's model, may be fortuitous they serve as useful guides for experimental investigations.

A. Lifshitz, Hebrew University, Israel. The induction time for the $COS + O_2 \rightarrow CO + SO_2$ reaction system has a very weak temperature dependence; ~ 17 Kcal/mole. Since the kinetics of these systems is very well known it is highly recommended for all cell size calculation by kinetic and hydrodynamic interaction. The system will only be weakly dependent on the exact temperature profile across the cell.

Author's Reply. This is an excellent proposal, since presumably for this system the hydrodynamic part of the interaction can be identified and validated before attempting to include more temperature sensitive chemical systems.

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) This paper reports on a series of field tests performed at the Canadian Defence Research Establishment Suffield and at Raufoss, Norway, to obtain the critical tube diameter, d_c , for ethylene-air mixtures by investigating the diffraction of detonations from tubes into large plastic bags simulating an unconfined fuel-air cloud. The critical ethylene-air compositions for successful re-establishment of detonation upon emerging from tubes of diameters 0.31 m, 0.45 m, 0.89 m and 1.36 m were determined by monitoring the diffracted detonation wave in the bag. High-speed cinematography of the diffracted wave shows that re-establishment of detonation is via one or more re-ignition centres at sites along the head of the expansion wave which originates at the area change. The characteristic transverse wave spac-		

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ings, S , associated with the detonations were measured from smoked foils mounted in the tubes. These measurements demonstrate that the empirical relation $d_c = 13 S$ provides a good correlation between the critical tube diameter and the cell size over a wide range of ethylene-air compositions. However, a better understanding of the coupled chemical-gas-dynamic processes within the cell is required in order to clarify the link between cell size and chemical kinetics.

In addition to the open tube tests, transmission of detonations through circular orifice holes was investigated in both ethylene-air and acetylene-air mixtures. Based on these tests it is concluded that the critical tube and critical orifice diameters are equal.

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APPENDIX V

DYNAMIC PARAMETERS OF GASEOUS DETONATIONS

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INTRODUCTION

In addition to gases, flammable liquids and solids in the form of fine droplets and dust particles also form explosive mixtures with air. An explosive mixture can, in general, support two modes of combustion. The slow laminar deflagration mode is at one extreme; here the flame propagates at typical velocities of the order 1 m s^{-1} relative to the unburned gases and there is negligible overpressure development when the explosion is unconfined. At the other extreme is the detonation mode, in which the detonation wave propagates at about 2000 m s^{-1} accompanied by an overpressure rise of about 20 bars across the wave. The propagation of laminar deflagrations is governed by the molecular diffusion of heat and mass from the reaction zone to the unburned mixture. The propagation of detonations depends on the adiabatic shock compression of the unburned mixtures to elevated temperatures to bring about autoignition. The very strong exponential temperature dependence of chemical reaction rates in general makes possible the rapid combustion in the detonation mode. Two-phase liquid droplets or dust-air mixtures are similar, but they require more physical processes (e.g. droplet break-up, phase change, mixing, etc.) prior to combustion. Thus, characteristic time or length scales associated with the combustion front are usually much larger than those of homogeneous gaseous fuel-air mixtures. The essential mechanisms of propagation of the combustion waves, however, are similar. In between the two extremes of laminar deflagration and detonation, there is an almost continuous spectrum of burning rates where turbulence plays the dominant role in the combustion process. Due to space limitations, only homogeneous gaseous fuel-air detonations are considered in this article.

The problem of fuel-air explosions has received considerable attention in recent years in connection with the safety aspects of large-scale transport and storage of liquified petroleum or natural gases. The Three Mile Island incident also raises the question of the possibility and consequences of a hydrogen-air explosion inside a nuclear-reactor containment. Two-phase liquid sprays and dust explosions are also of concern in accidental fuel release in petrochemical plants and in grain elevators and coal mines. As a result of the current worldwide interest in fuel-air explosions, active research involving large-scale field tests has been carried out in recent years. In this paper, we single out the gaseous fuel-air detonation part of fuel-air explosions, reporting on the progress made and the current state of the art.

Accidental fuel-air explosions are discussed in the recent monographs of Gulan (1978), Bartknecht (1978), and Baker et al. (1983), as well as in an excellent general review article by Strehlow (1980). In addition, the proceedings of a recent specialist meeting on fuel-air explosions contains progress reports of current activities at various laboratories around the world (Lee & Guirao 1982). The present paper, therefore, avoids duplication of material that can be found elsewhere. Efforts are made, however, to keep the paper self-contained.

THE DYNAMIC PARAMETERS OF DETONATIONS

The classical Chapman-Jouguet theory, in essence, seeks the unique solution of the one-dimensional conservation equations across the detonation front in which the flow behind the wave is sonic. It does not require a knowledge of the structure of the wave itself, and involves only an equilibrium thermodynamic calculation for the detonation states (i.e. the detonation velocity, pressure, temperature, and density ratios across the wave, and the equilibrium composition of the product gases). The equilibrium, or "static," detonation states obtained from the classical Chapman-Jouguet theory agree surprisingly well with experimental observations, even in near-limit conditions when the flow structure near the front is highly three-dimensional.

The parameters requiring a knowledge of the structure, and hence the chemical reaction rates, are the detonability limits, the initiation energy, the critical tube diameter, and the thickness of the reaction zone. We henceforth refer to these parameters as the dynamic detonation parameters to distinguish them from the equilibrium "static" detonation states obtained from the Chapman-Jouguet theory. A one-dimensional model for the detonation structure was proposed in the early 1940s by Ya. B. Zel'dovich, W. Döring, and J. von Neumann (i.e. the ZDN model). It assumes a planar shock followed by a reaction zone after an induction delay period. This

model permits the computation of the dynamic parameters when a model for the physical processes involved is given. However, results from such computations using the ZDN model for the detonation structure are quite far from those obtained from experiments, mainly because the ZDN structure is unstable and is never observed experimentally except under transient conditions. This is in accord with experimental observations that all self-sustained detonations have a three-dimensional cellular structure. A quantitative theory for the real cellular detonation structure has yet to be developed. Thus, it seems rather surprising that a century after the detonation phenomenon was first identified, and over eighty years since the successful Chapman-Jouguet theory for the prediction of the equilibrium properties was proposed, there is still no quantitative theory for estimating these dynamic detonation parameters. They remain as experimental parameters, and their measurements are not without some fundamental difficulties either.

Detonation research up until the early 1970s was mostly aimed at furthering our understanding of the physical and chemical processes involved in the various aspects of the detonation phenomena (i.e. transition from deflagration to detonation, onset of detonation, blast initiation, shock interaction processes leading to the so called multiheaded or cellular structure, etc.). Thus, sensitive mixtures of fuels (e.g. H_2 , C_2H_2) with pure oxygen were used in general, and the experiments were usually carried out at subatmospheric initial pressures for convenience. Measurements of the dynamic parameters were also mostly made on fuel-oxygen mixtures at subatmospheric initial pressures, since the objective of these measurements was to study the dependence of these dynamic detonation parameters on the other experimental variables. As a result, there are very few experimental data on these dynamic parameters, particularly on practical systems, to permit even empirical correlations to be made.

In Volume 5 of this series, Oppenheim & Soloukhin (1973) gave a very comprehensive historical summary of detonation research up to that time, as well as an extensive bibliography of the early works. Of particular interest in their article are the beautiful laser-schlieren cinematographic records of the various physical processes associated with the phenomena of transition and initiation, as well as the detailed cellular structure of the detonation front itself. These exceptional experimental records provide a good qualitative understanding of the detonation phenomena, and readers are encouraged to refer to this article for a physical background on the subject. Other noteworthy review articles summarizing the detailed structure of cellular detonations are those of Edwards (1969) and Strehlow (1968). The recent monograph by Fickett & Davis (1979) also gives a good comprehensive review of the stability and structure of cellular detonations.

More recent reviews emphasizing the direct initiation and the transition from deflagration to detonation have been given by Lee (1977) and Lee & Moen (1980).

It may be concluded that by the early 1970s all aspects of the detonation phenomena were quite well understood, at least on a qualitative basis. There is, however, a strong deficiency in reliable experimental data on the dynamic detonation parameters, particularly in fuel-air mixtures at atmospheric initial pressures that are of practical interest. There are also very few results on fuel-air detonations due to the fact that very-high-energy sources such as solid explosive charges must be used for the initiation as a result of their relative insensitivity. Also, the size of the apparatus required necessitates that the experiments be carried out in the field. However, as a result of the current interest in accidental fuel-air explosions, a number of large-scale experiments have been carried out in the past decade on fuel-air detonations. We emphasize here the discussion of these new results and their correlations, and point out the progress made toward the prediction of the dynamic detonation parameters.

THE DETONATION CELL SIZE λ

Perhaps the most important dynamic parameter is the average size λ of a cell of the detonation front. Because of its fundamental significance, it is worthwhile to review briefly the essential features of a cellular detonation front. Figure 1 shows the pattern made by the normal reflection of a detonation on a glass plate coated lightly with carbon soot, which may be from either a wooden match or a kerosene lamp. The cellular structure of the detonation front is quite evident. If a similarly soot-coated polished metal (or mylar) foil is inserted into a detonation tube, the passage of the detonation wave will leave a characteristic "fish-scale" pattern on the smoked foil. Figure 2 is a sequence of laser-schlieren records of a detonation wave propagating in a rectangular tube. One of the side windows has been coated with smoke, and the fish scale pattern formed by the propagating detonation front is very well illustrated. The detailed shock configuration of the cellular detonation front itself is illustrated by the interferogram shown in Figure 3. The direction of propagation of the detonation is toward the right. As can be seen in the sketch at the top left corner, there are two triple points. At the first triple point A, AI and AM represent the incident shock and Mach stem of the leading front, while AB is the reflected shock. Point B is the second triple point of another three-shock Mach configuration on the reflected shock AB; the entire shock pattern represents what is generally referred to as a double Mach reflection. The hatched lines denote the reaction front, while the dash-dot lines represent the shear discontinuities

or slip lines associated with the triple-shock Mach configurations. The entire front ABCDE is generally referred to as the transverse wave, and it propagates normal to the direction of the detonation motion (down in the present case) at about the sound speed C_1 of the hot product gases. It has been shown conclusively that it is the triple-point regions at A and B that "write" on the smoked foil. The exact mechanics of how the triple-point region does the writing is not clear. It has been postulated that the high shear at the slip discontinuity causes the soot particles to be erased. Figure 4 shows a schematic of the motion of the detonation front. The fish-scale pattern is a record of the trajectories of the triple points. It is important to note the cyclic motion of the detonation front. Starting at the apex of the cell at A, the detonation shock front is highly overdriven, propagating at about

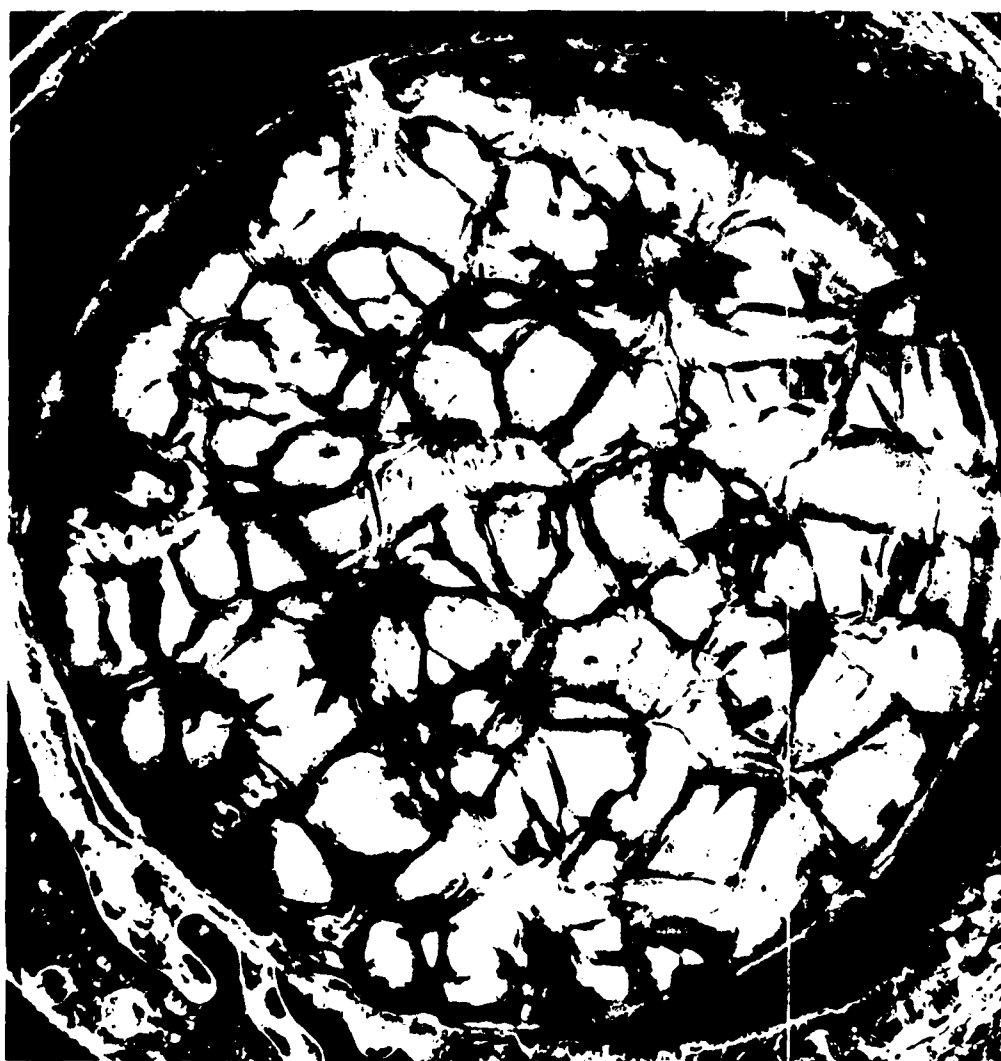


Figure 1 End-on pattern from the normal reflection of a cellular detonation on a smoked glass plate.

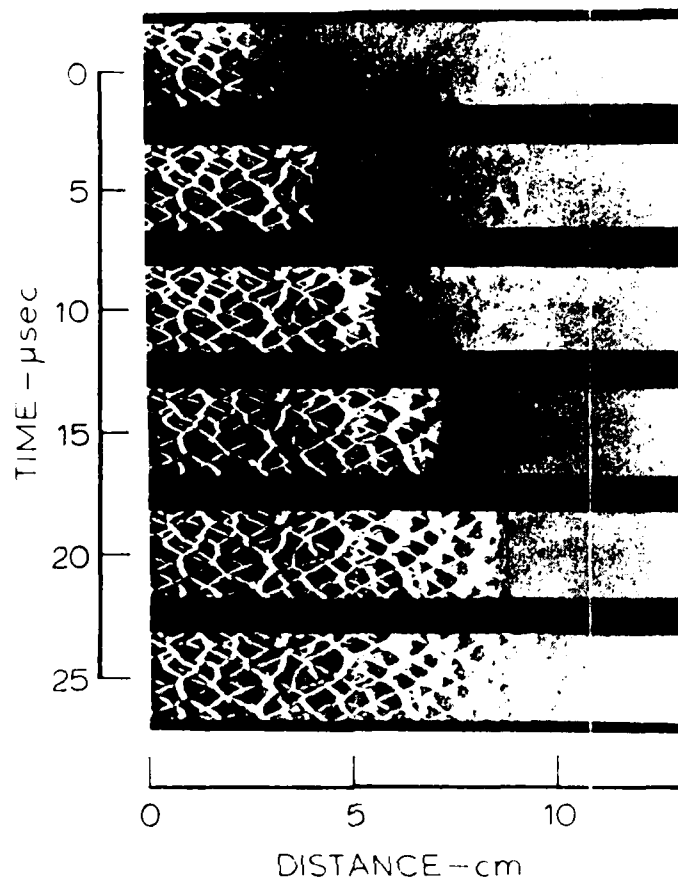


Figure 2 Laser-schlieren cinematography of a propagating detonation in low-pressure H_2-O_2 mixtures, with fish-scale pattern shown on a soot-coated window (courtesy of A. K. Oppenheim).

1.6 times the equilibrium Chapman-Jouguet detonation velocity. Toward the end of the cell at D, the shock has decayed to about 0.6 times the Chapman-Jouguet velocity before it is impulsively accelerated back to its highly overdriven state when the transverse waves collide to start the next cycle again. For the first half of the propagation from A to BC, the wave serves as the Mach stem to the incident shocks of the adjacent cells. During the second half from BC to D, the wave then becomes the incident shock to the Mach stems of the neighboring cells. Details of the variation of the shock strength and chemical reactions inside a cell can be found in a recent paper by Libouton et al. (1981). AD is usually defined as the length L_c of the cell, and BC denotes the cell diameter (also referred to as the cell width or the transverse-wave spacing). The average velocity of the wave is close to the equilibrium Chapman-Jouguet velocity.

We thus see that the motion of a real detonation front is far from the steady and one-dimensional motion given by the ZDN model. Instead, it proceeds in a cyclic manner in which the shock velocity fluctuates within a

cell about the equilibrium Chapman-Jouguet value. Chemical reactions are essentially complete within a cycle or a cell length. However, the gasdynamic flow structure is highly three-dimensional, and full equilibration of the transverse shocks, so that the flow becomes essentially one-dimensional, will probably take an additional distance of the order of a few more cell lengths.

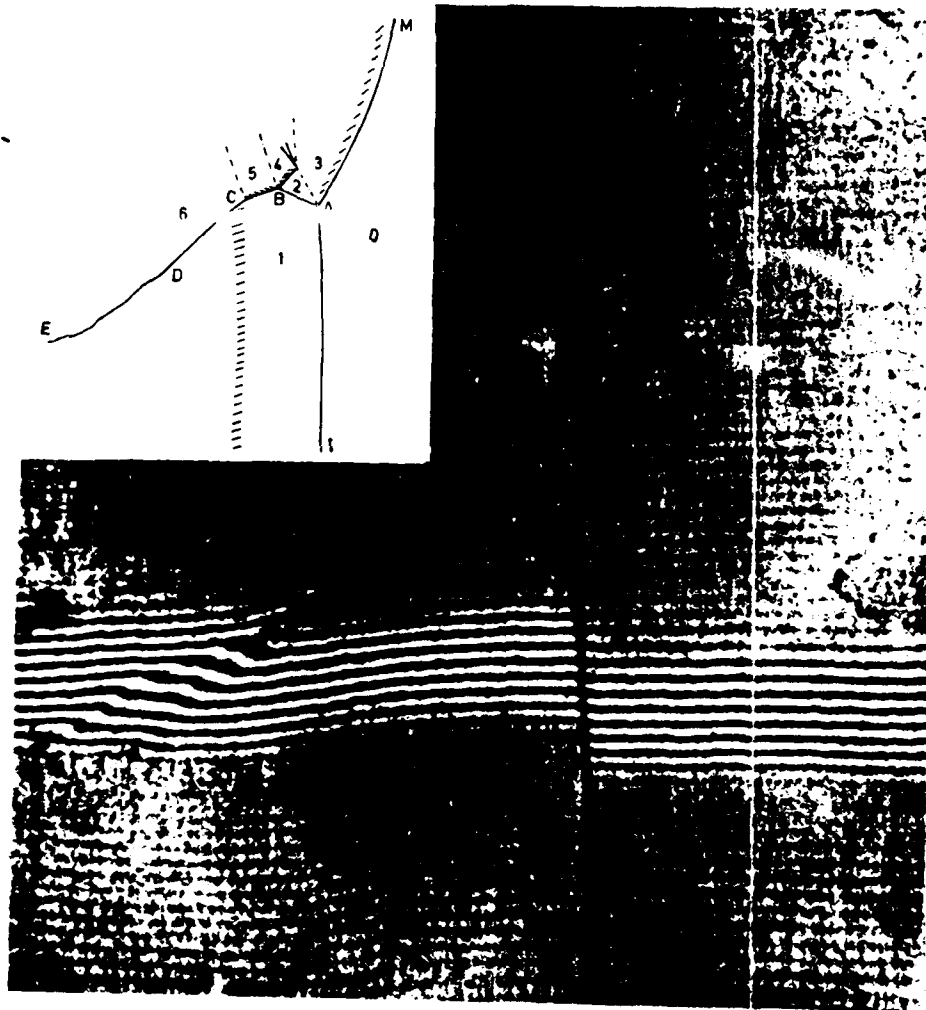


Figure 3 Interferogram of the detailed double Mach-reflection configuration of the structure of a cellular detonation front (courtesy of D. H. Edwards).

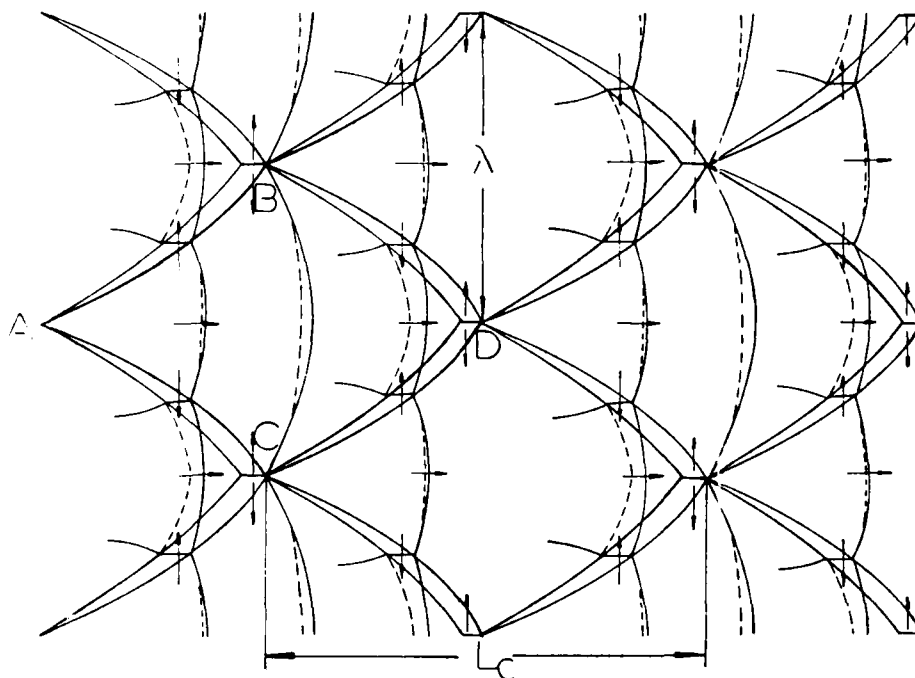


Figure 4 Schematic of the propagation of a cellular detonation front, showing the trajectories of the triple points.

From both the cellular end-on or the axial fish-scale smoke foil, the average cell size λ can be measured. The end-on record gives the cellular pattern at one precise instant. The axial record, however, permits the detonation to be observed as it travels along the length of the foil. It is much easier by far to pick out the characteristic cell size λ from the axial record; thus, the end-on pattern is not used, in general, for cell-size measurements.

Early measurements of the cell size have been carried out mostly in low-pressure fuel-oxygen mixtures diluted with inert gases such as He, Ar, and N_2 (Strehlow & Engel 1969). The purpose of these investigations is to explore the details of the detonation structure and to find out the factors that control it. It was not until very recently that Bull et al. (1982) made some cell-size measurements in stoichiometric fuel-air mixtures at atmospheric pressure. Due to the fundamental importance of the cell size in the correlation with the other dynamic parameters, a systematic program has been carried out by Knystautas to measure the cell size of atmospheric fuel-air detonations in all the common fuels (e.g. H_2 , C_2H_2 , C_2H_4 , C_3H_6 , C_2H_6 , C_3H_8 , C_4H_{10} , and the welding fuel MAPP) over the entire range of fuel composition between the limits (Knystautas et al. 1983). Stoichiometric mixtures of these fuels with pure oxygen, and with varying degrees of N_2 dilution at atmospheric pressures, were also studied (Knystautas et al. 1982). To investigate the pressure dependence, Knystautas et al. (1982) have

also measured the cell size in a variety of stoichiometric fuel-oxygen mixtures at initial pressures $10 \lesssim p_0 \lesssim 200$ torr. Figure 5 gives the typical U-shaped curves of the variation of the cell size λ for some of the common hydrocarbon fuel-air mixtures with the equivalence ratio ϕ (i.e. $\phi = 1$ for stoichiometric composition) of the mixture. The minimum cell size usually occurs at about the most detonable composition ($\phi = 1$). The cell size λ is representative of the sensitivity of the mixture. Thus, in descending order of

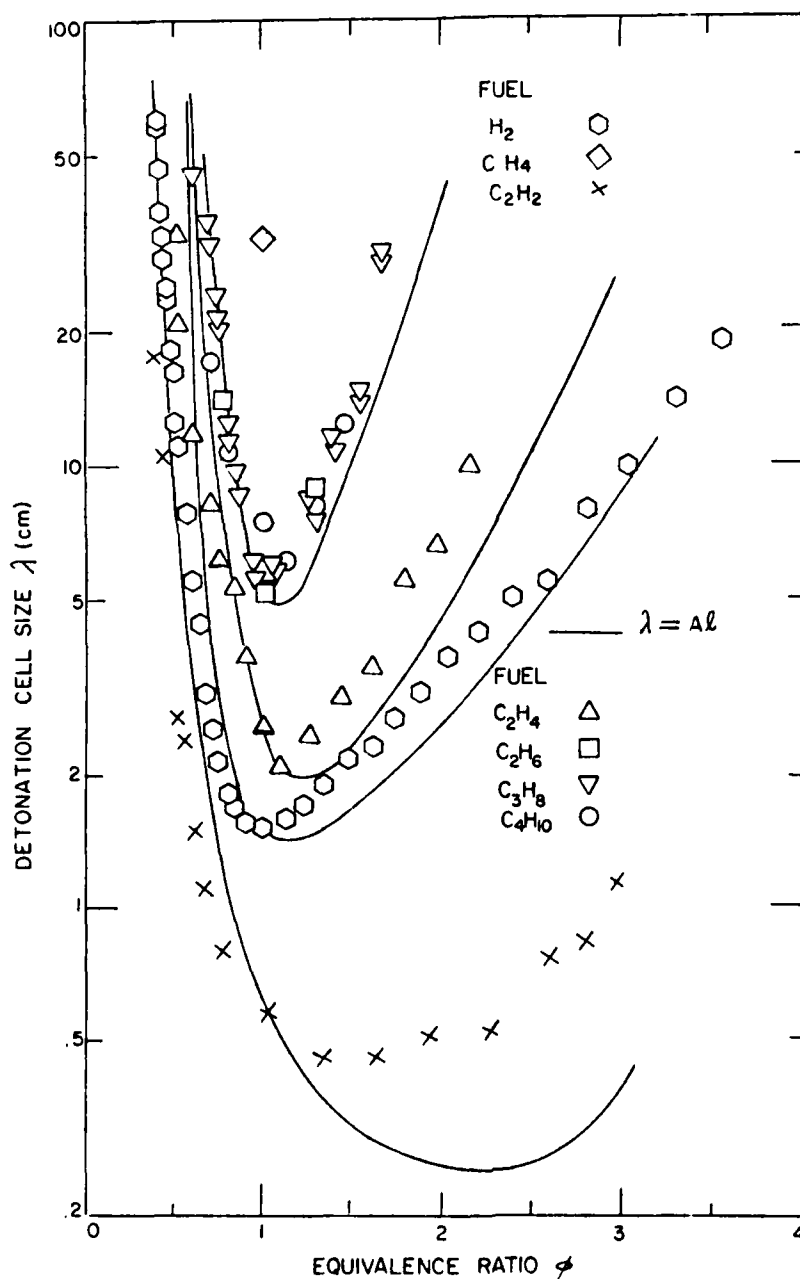


Figure 5 Cell size of fuel-air mixtures at atmospheric pressure.

sensitivity, we have C_2H_2 , H_2 , C_2H_4 , and the alkanes C_3H_8 , C_2H_6 , and C_4H_{10} . Methane (CH_4), although belonging to the same alkane family, is exceptionally insensitive to detonation, with an estimated cell size $\lambda \approx 33$ cm for stoichiometric composition as compared with the corresponding value of $\lambda \approx 5.35$ cm for the other alkanes. That the cell size λ is proportional to the induction time of the mixture had been suggested by Shchelkin & Troshin (1965) long ago. However, to compute an induction time requires that the model for the detonation structure be known, and no theory exists as yet for the real three-dimensional structure. Nevertheless, one can use the classical ZDN model for the structure and compute an induction time or, equivalently, an induction-zone length l . While this is not expected to correspond to the cell size λ (or cell length L_c), it may elucidate the dependence of λ on l itself (e.g. a linear dependence $\lambda = Al$ as suggested by Shchelkin & Troshin 1965). Westbrook (1982, Westbrook & Urtiew 1982) has made computations of the induction-zone length l using the ZDN model, but his calculations are based on a constant-volume process after the shock, rather than integration along the Rayleigh line. Very detailed kinetics of the oxidation processes are employed. By matching with one experimental point, the proportionality constant A can be obtained. This is done in Figure 5 by using Westbrook's data for l and matching at $\phi = 1$. The solid lines in the figure represent the linear relationship $\lambda = Al$, where l is computed from detailed kinetics. The general dependence of λ on ϕ is reproduced qualitatively, but quantitatively the agreement is not as good as it appears because errors are hidden by the very steep nature of the U-shaped curve. The agreement, however, is within one order of magnitude. The constant A differs for different gas mixtures (e.g. $A = 10.14$ for C_2H_4 , $A = 52.23$ for H_2); thus, the three-dimensional gasdynamic processes cannot be represented by a single constant alone over a range of fuel composition for all the mixtures. The chemical reactions in a detonation wave are strongly coupled to the details of the transient gasdynamic processes, with the end product of the coupling being manifested by a characteristic chemical length scale λ (or equivalently L_c) or time scale $t_c = \lambda/C_1$ (where C_1 denotes the sound speed in the product gases, which is approximately the velocity of the transverse waves) that describes the global rate of the chemical reactions. Since $\lambda \approx 0.6L_c$ and $C_1 \approx 0.5D$, where D is the Chapman-Jouguet detonation velocity, we have $t_c \approx L_c/D$, which corresponds to the fact that the chemical reactions are essentially completed within one cell length (or one cycle).

It appears that a correct model for the cellular structure must be a time-dependent one in which the nonlinear coupling mechanism between gasdynamics and chemical kinetics can be properly modeled. Computer codes for transient reacting flows with shock waves are currently available.

The feasibility of modeling the real cellular structure of a two-dimensional detonation front has also been demonstrated recently by Taki & Fujiwara (1981) and Oran et al. (1981). However, an extension to three dimensions with the appropriate spatial resolutions to account for the details of the complex double Mach configurations, although possible in principle, would require computer time and storage capacity much in excess of what even the current fast machines are capable of. Thus, the use of numerical simulation as a tool for obtaining cell-size data would not be practical, and direct experimental measurement remains as the most convenient means of determining the cell size λ . Although other methods have been tried to measure the cell size directly, the simple smoke-foil technique has not been improved upon to date and still remains as the only successful method. For near-limit fuel-air detonations, where the cells are very large and foils on the order of meters in length have to be used, the actual deposition of a uniform coating of soot from numerous burners poses a rather difficult engineering problem. A more serious problem in the measurement of cell size is the actual interpretation of the fish-scale pattern. For most fuel-air mixtures, this pattern is highly irregular, and thus the selection of the "correct" cell size requires a certain amount of experience. This introduces a subjective element into the measurement of λ , which has to be resolved. The use of long foils so that the wave is recorded over a long travel length certainly facilitates the interpretation of the foil. Alternatively, if a large number of experiments are made in order to accumulate an ensemble of records under identical conditions, then this also improves the accuracy of the measurement. However, other techniques must be developed to facilitate the measurement of this important fundamental dynamic parameter, which characterizes the real structure of the detonation wave.

THE CRITICAL TUBE DIAMETER

Another important dynamic parameter that has received considerable attention in recent years is the so-called critical tube diameter. Experimentally, it is found that if a planar detonation wave propagating in a circular tube emerges suddenly into an unconfined volume containing the same mixture, the planar wave will transform into a spherical wave if the tube diameter d exceeds a certain critical value d_c (i.e. $d \geq d_c$). If $d < d_c$, the expansion waves will decouple the reaction zone from the shock, and a spherical deflagration wave results. Excellent schlieren records of these transmission phenomena for $d \geq d_c$ and $d < d_c$ can be found in the review by Oppenheim & Soloukhin (1973).

Perhaps the most important progress in recent years has been in the linking together of the various dynamic parameters. This work originates

from the demonstration of a universal correlation between the critical tube diameter and the cell size, an observation first made some 20 years ago by Mitrofanov & Soloukhin (1965). In studying the diffraction of a planar detonation wave as it emerges from a circular tube into unconfined space containing the same mixture, they noticed that the critical tube diameter is about 13 times the cell size of the mixture (i.e. $d_c = 13\lambda$). For a square tube, the critical width W_c is found to be of the order of 10 times the cell size (i.e. $W_c = 10\lambda$). Since they based their observation on one particular mixture of stoichiometric acetylene and oxygen over a narrow range of initial pressures (i.e. $p_0 \lesssim 100$ torr), the significance of this observation was not recognized. Over a decade elapsed before Edwards et al. (1979) brought up this observation again. In repeating Mitrofanov & Soloukhin's experiments, they confirmed the earlier result that $d_c = 13\lambda$. Furthermore, they also pointed out that there is no reason why this correlation should not be applicable to all other detonative systems. Moen et al. (1981) followed up on this suggestion and demonstrated the validity of the 13λ correlation for stoichiometric ethylene-oxygen mixtures with nitrogen dilution. A systematic experimental program was then carried out by Knystautas et al., in which cell sizes and critical tube diameters were simultaneously measured for all the common hydrocarbons (i.e. H_2 , C_2H_2 , C_2H_4 , C_3H_6 , C_2H_6 , C_3H_8 , CH_4 , and MAPP). Cell sizes were measured in stoichiometric mixtures of these fuels with pure oxygen at low initial pressures ($p_0 \lesssim 150$ torr), as well as in stoichiometric fuel-oxygen mixtures at atmospheric pressure, but with different degrees of nitrogen dilution. A comparison with the direct measurements of the critical tube diameter confirmed the universality of the 13λ correlation. A comprehensive report of the detailed results of this important study can be found in Knystautas et al. (1981). The cell-size measurement has recently been further extended to fuel-air mixtures over the range of fuel concentration between the limits. We have seen some of these cell-size data already in Figure 5. There have also been a number of large-scale field experiments on the direct measurements of the critical tube diameter in fuel-air mixtures (Rinnan 1982, Guirao et al. 1982, Moen et al. 1983). The results are summarized in Figure 6, which shows the dependence of the critical tube diameter d_c on the equivalence ratio ϕ (i.e. mixture composition) for some of these common fuel-air mixtures. The solid curves are the cell-size data from Figure 5 multiplied by 13. As can be observed, the agreement is sufficiently good to confirm the universal nature of the $d_c = 13\lambda$ correlation. Some doubt has been expressed by Moen et al. (1983), who simultaneously measured both cell size and critical tube diameter. They reported that, based on their own cell-size data, $14\lambda \lesssim d_c \lesssim 24\lambda$. However, their critical tube diameter data agree perfectly with the 13λ correlation when the cell-size data obtained independently by Knystautas

et al. (1983) are used. Knystautas et al's cell-size data are judged more reliable in that very long smoke foils were used and usually a number of records for the same mixture were taken to ensure a correct interpretation of the true cell size. Furthermore, independent estimates of the cell size have also been obtained from the frequencies of the pressure oscillations from the pressure profile recorded by the transducers mounted on the detonation tube. An assessment of the data available thus far indicates that $d_c = 13\lambda$ is indeed a general relationship, at least for the range of fuels tested. It should be pointed out that the $d_c = 13\lambda$ correlation is extremely useful in practice. The cell size λ can be determined relatively easily from laboratory-scale

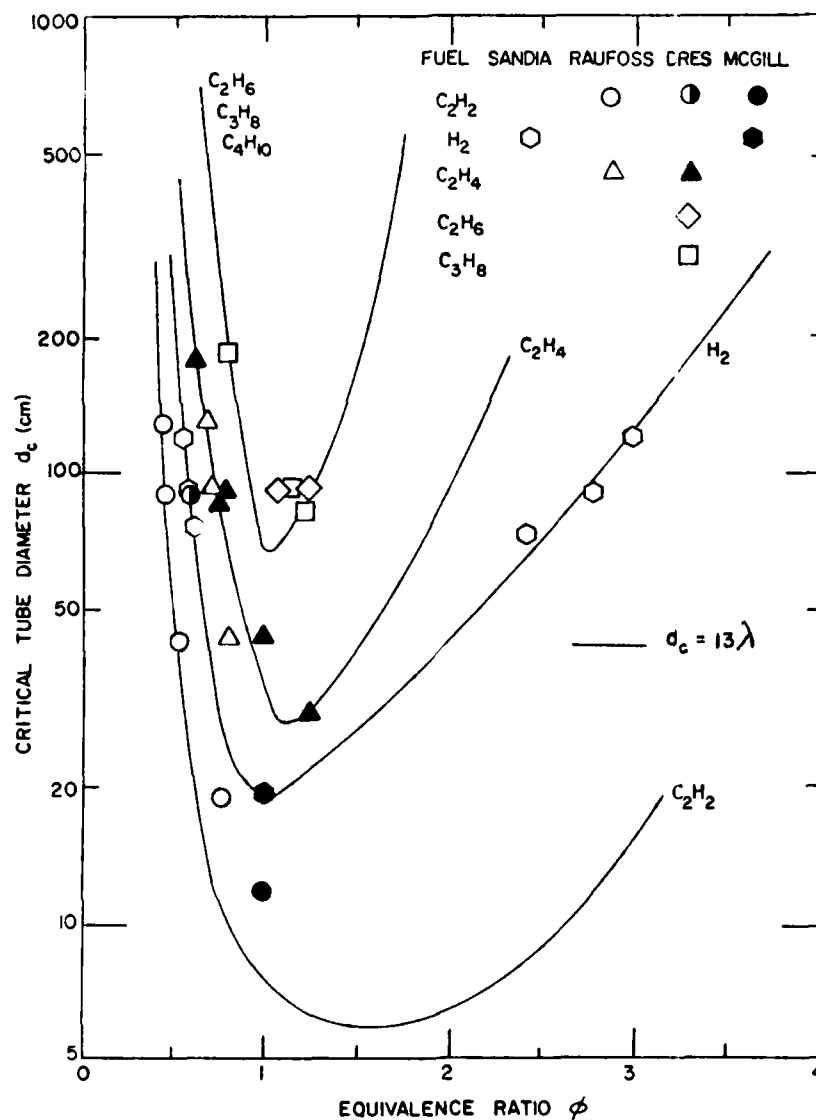


Figure 6 Comparison of experimental data for the critical tube diameter in fuel-air mixtures with the 13λ correlation.

detonation tubes under easily controlled conditions, thus enabling the critical tube diameter to be obtained immediately. Direct measurements of the critical tube diameter require rather large-scale field experiments, in general, for the relatively insensitive fuel-air mixtures.

There are no quantitative theories for the prediction of the critical tube diameter as yet. From physical considerations, a qualitative criterion for the transmission can be developed. When the detonation emerges from the tube, rarefaction waves are generated at the circumference that propagate toward the tube axis as the high-pressure detonation products expand radially outwards. These waves cool the shocked gases and thus increase the induction time, causing the reaction zone to decouple from the leading shock front. The characteristic time for this gasdynamic quenching process will be of the order of the tube radius divided by the sound speed of the product gases (i.e. $t_c = R_c/C_1$). It seems reasonable to assume that if the effective thickness of the detonation wave (i.e. from the leading shock to the equilibrium Chapman-Jouguet plane) is Δ_H , then the detonation must propagate a distance of at least $2\Delta_H$ prior to the rarefaction waves reaching the tube axis for it not to be quenched. In this way, at least a small detonation core near the tube axis is not influenced by the gasdynamic quenching, and this core serves as a kernel that subsequently develops into a spherical wave. The time t_D for the detonation to propagate an axial distance of $2\Delta_H$ will simply be $2\Delta_H/D$, where D is the Chapman-Jouguet velocity. Equating the two characteristic times (i.e. $t_D = t_c$) gives $R_c = 2C_1\Delta_H/D$. Since, for most detonable mixtures, the detonation velocity $D \simeq 2C_1$, we see that the above criterion gives $R_c \simeq \Delta_H$ or the critical tube diameter $d_c \simeq 2\Delta_H$. The effective thickness, or the so-called hydrodynamic thickness, of a detonation wave is reported by Edwards et al. (1976) to be between 2.5 to 4 cell lengths, or 5 to 8 cell diameters. They base this result on measurements of the decay of the transverse pressure vibrations (i.e. equilibration of the transverse shocks) in oxyhydrogen and oxyacetylene mixtures. Their results are in accord with earlier independent measurements by Vasiliev et al. (1972). Taking an averaged value of Δ_H from Edwards et al's observations (i.e. $\Delta_H \simeq 6.5\lambda$), we see that the proposed criterion ($t_c/t_D = 1$) leads to the result $d_c \simeq 2\Delta_H = 13\lambda$, which is in perfect agreement with experiment. From the above discussions, the critical-tube-diameter problem is thus reduced to the resolution of the hydrodynamic or effective thickness of the cellular detonation front. Knowledge of Δ_H permits d_c to be determined.

The recent experimental work of Liu et al. (1983) has also produced some interesting results that shed further light on the critical-tube-diameter problem. In studying the transmission through an orifice plate, instead of from a straight tube, it is found that the critical orifice diameter is identical

to the critical tube diameter. Thus, the upstream pressure enhancement from the reflected detonation off the orifice plate does not influence the transition process from a planar to a spherical wave. The reflected detonation generates a radially converging shock wave, which, like the rarefaction waves, propagates toward the tube axis at about the sound speed of the product gases as a result of the high temperatures (and, hence, weak shock). The fact that there is no difference between the corresponding critical orifice and the critical tube diameters suggests that the unattenuated core near the tube axis, rather than the details of the exact gasdynamic flow structure behind the diffracted wave, dominates the development of the spherical detonation.

Experiments on the transmission through different-shaped orifice plates (i.e. square, rectangular, triangular, and elliptical) indicate that the 13λ correlation still holds if an effective diameter d_{eff} , which is the mean value of the largest and smallest dimensions that characterize the orifice opening, is used (i.e. $d_{\text{eff}} = 13\lambda$). For a square orifice of side length W , the smallest characteristic dimension would be W , while the largest would be the diagonal $\sqrt{2}W$. Thus, the effective diameter of the square orifice would be $d_{\text{eff}} = 1/2(W + \sqrt{2}W) \simeq 1.2W$. Hence, $d_{\text{eff}} = 13\lambda$ gives $W_c = (13/1.2)\lambda \simeq 10.8\lambda$, in close agreement with the earlier observations of $W_c = 10\lambda$ by Mitrofanov & Soloukhin (1965) and Edwards et al. (1979). The effective diameter is also the mean value of the diameter of the circumscribed and inscribed circles of the orifice opening. It would appear that the smallest dimension should be more important, since it controls the time for the rarefaction to penetrate the wave. However, when the two characteristic dimensions are not too different, the expansion is three-dimensional, and thus the mean value would be more appropriate to characterize the gasdynamic process.

In generalizing the results, one could also introduce the concept of wave curvature. The critical condition for transmission could then be stated in terms of a minimum radius of curvature of the diffracted wave. If the rarefaction waves give rise to a curvature of the diffracted detonation exceeding a certain critical value, then failure of the wave results. The maximum curvature (or minimum radius of curvature R) would be of the order of about the hydrodynamic thickness Δ_H (i.e. $R = \Delta_H = 6.5\lambda$) for the three-dimensional case of the transformation from arbitrarily shaped planar to spherical geometry.

For rectangular orifices (or slots) where the aspect ratio L/W becomes large (e.g. $L/W \gtrsim 7$) and the planar wave transforms to a cylindrical wave, Liu et al. (1983) found that the smaller length W now controls the transmission process, rather than the mean value of the two characteristic dimensions L and W . Based on the wave-curvature concept, the cor-

responding radius of curvature of a two-dimensional cylindrical wave would be one half that of the spherical wave for the same curvature. Thus, if $R = \Delta_H$ for the three-dimensional case, $R = \Delta_H/2 = 3.25\lambda$ for the two dimensional case. This is in agreement with the experimental result of Liu that $W_c \approx 3.25\lambda$.

Since Liu's experiments are based on rectangular slot orifices of $L/W \lesssim 7$, some large-scale experiments on two-dimensional rectangular channels with $L/W \approx 35$ have been carried out recently by Benedick et al. (1983) to verify the $W_c \approx 3\lambda$ limit. Their experiments used H_2 -air and C_2H_4 -air mixtures at atmospheric pressures, instead of the H_2 - O_2 - N_2 and C_2H_4 - O_2 - N_2 mixtures used by Liu. The results, however, confirmed the two-dimensional limit of $W_c = 3\lambda$ found by Liu. Furthermore, a half-channel experiment was also performed that demonstrated symmetry about the centerline, in that a value of $W_c/2 = 1.5\lambda$ was obtained.

These very recent experimental findings are extremely interesting. It is clear that the critical-tube-diameter problem provides much insight into the coupling mechanisms between gasdynamics and chemical kinetics. The sudden emergence from confined to unconfined space, in essence, subjects the detonation to a finite perturbation. Whether it survives and readjusts to the new geometry or fails will define the minimum requirements for the self-sustained propagation of the wave under its new environment. The recent study of Murray & Lee (1983) on detonation failure when the detonation transmits from a rigid steel tube to a thin plastic tube is of particular interest in this respect. By using plastic tubes of different wall thickness, the severity of the gasdynamic expansion can be controlled, and the failure criterion in this case can then be related to the wave curvature. It should be noted that even in rigid tubes, the negative displacement thickness of the boundary layer causes flow divergence much like the yielding plastic wall experiment of Murray & Lee (1983). Thus, rarefaction waves are generated at the walls, which then propagate toward the tube axis and cause the wave to be curved. Direct measurements of the radius of curvature of a detonation wave have been carried out recently by Desbordes et al. (1981). They reported a radius of curvature of about 1 m for a planar detonation in a 10-cm-diameter tube. The corresponding curvature of the wave is much smaller than the critical value for the failure of a spherical wave (i.e. $1/R \approx 1/6.5\lambda$). Thus, it appears that the minimum radius of curvature of a spherical wave would be about the hydrodynamic thickness $\Delta_H \approx 3.5L_c \approx 6.5\lambda$. For waves with radii of curvature much larger than the critical value, a velocity deficit results. Thus, failure in rigid and plastic tubes, as well as sudden expansion into unconfined space, are all similar processes and differ only by the degree of the transverse expansion itself. Since the lateral expansion results in a curved front, the propagation criterion under different boundary conditions can be unified under the wave-curvature concept.

CRITICAL ENERGY FOR DIRECT INITIATION

Experimentally, it is found that for a given mixture at given initial conditions, a definite quantity of energy must be used to initiate a detonation "instantaneously." By "instantaneously" is meant that the initial strong blast wave generated by the powerful igniter upon the rapid deposition of its energy decays asymptotically to a Chapman-Jouguet detonation. If the igniter energy is less than a certain critical value, the reaction zone progressively decouples from the blast as it decays and a deflagration results. Since the transition from deflagration to detonation under unconfined conditions is extremely difficult, if even possible, spherical detonations are almost always initiated in practice via the direct or blast-initiation mode. Since the subject of direct initiation has been thoroughly reviewed previously (Lee 1977), it suffices to consider only the recent developments and results.

The interest in unconfined fuel-air detonations in recent years has led to a number of experimental studies on the measurement of the critical initiation energy. The most extensive of these measurements for fuel-air mixtures are those by Bull et al. (1978). The dependence of the critical energy (or, equivalently, the weight of the solid explosive charge used) on the equivalence ratio ϕ is, like all dynamic parameters, in the form of U-shaped curves with the minimum around stoichiometric composition (i.e. $\phi = 1$). The minimum energy at $\phi = 1$ has been used as a relative measure of the sensitivity of the various fuels to detonation (Matsui & Lee 1978). Also, by specifying somewhat arbitrarily an upper limit of the critical energy above which the mixture is rendered nondetonable, the U-shaped curve can be used to define the detonability limits for unconfined detonations. Although there are a number of experimental parameters that may influence the initiation energy (e.g. the type of igniter, its geometry, energy-time characteristics, etc.), it can be concluded that as long as the time for the energy deposition is short compared with the characteristic time of the blast wave [i.e. R_0/C_0 , where $R_0 = (E_0/p_0)^{1/3}$ is the explosion length and C_0 is the sound speed], the blast wave can be considered as an ideal point spherical blast characterized by the total energy E_0 only. Thus, experimental measurements of the critical energy in fuel-air mixtures using concentrated solid explosive charges satisfy this condition, and we need not consider other details of the source itself.

The various theories for the critical energy have already been reviewed (Lee 1977). They all show a cubic dependence of the critical energy E_0 on the induction time τ for spherical detonations (i.e. $E_0 \sim \tau^3$). However, the lack of an overall chemical length or time that characterizes the real three-dimensional cellular structure of the detonation wave prevents all these theoretical efforts from being completely quantitative. The use of an

experimental data point is usually necessary to evaluate a certain constant of proportionality between E_0 and τ^3 . As an example, consider the simplest theory, based on Zel'dovich's criterion (see Lee 1977) that when the initiating blast has decayed to the Chapman-Jouguet strength, the decay time must be of the order of the induction time. Using strong-blast theory, Lee (1977) derived the following expression for the critical energy E_0 :

$$E_0 = k_j \rho_0 I D^{j+3} [(j+3)/2]^{j+1} \tau^{j+1}, \quad (1)$$

where $k_j = 1, 2\pi, 4\pi$ for $j = 0, 1, 2$, corresponding to the planar, cylindrical, and spherical geometry, respectively; I is a numerical constant that is a function of the specific heat ratio γ (for $\gamma = 1.4$, $I = 0.423, 0.626, 1.212$ for $j = 2, 1, 0$, respectively); ρ_0 is the initial density; D is the Chapman-Jouguet velocity, and τ is a characteristic chemical time. If the induction time evaluated at the shock temperature corresponding to a Chapman-Jouguet detonation is used for τ , Equation (1) gives values for E_0 about three orders of magnitude smaller than the experimental values. A more appropriate overall chemical time can be derived from the cell length, which is approximately twice the cell diameter λ , because chemical reactions are essentially complete within one cell length. Thus, we may define

$$\tau_c = 2\lambda/D,$$

and Equation 1 then becomes

$$E_0 = k_j \rho_0 I (j+3)^{j+1} D^2 \lambda^{j+1}, \quad (2a)$$

$$E_0 = 500\pi \rho_0 I D^2 \lambda^3 \quad \text{for } j = 2. \quad (2b)$$

A knowledge of the cell diameter λ from experiment permits the evaluation of the critical energy E_0 . The Chapman-Jouguet velocity D and the value for the constant I can be determined easily when the initial state and the mixture composition are specified.

Another simple relationship linking the cell diameter to the critical energy can be obtained via the critical tube diameter. Based on the work-done concept, Lee & Matsui (1977) derived a simple expression for E_0 and d_c as

$$E_0 = \frac{\pi p_1 u_1}{24 c_1} d_c^3, \quad (3)$$

where p_1 , u_1 , and c_1 are the pressure, particle velocity, and sound speed at the Chapman-Jouguet plane. Using the Rankine-Hugoniot conditions across a Chapman-Jouguet detonation, Equation (3) may also be written as

$$E_0 = \frac{\pi \rho_0 D^2}{24 \gamma (\gamma + 1)} d_c^3 = \frac{2197 \pi \rho_0 D^2 \lambda^3}{24 \gamma (\gamma + 1)} \quad (4)$$

when the 13λ correlation is used for d_c .

Recently, Lee et al. (1982) have proposed a more direct link between the initiation energy and the cell size via the critical tube diameter. The idea is based on the requirement of a minimum surface energy before a planar wave can evolve into a spherical wave without failure. This minimum surface energy should correspond to the area of the critical tube, $\pi d_c^2/4$. Thus, in blast initiation, the blast energy E_0 must be such that when the wave has decayed to the Chapman-Jouguet strength, the surface energy of the blast sphere must be at least proportional to $\pi d_c^2/4$. Equating directly the surface area of the blast sphere to the area of the critical tube, we write

$$4\pi R_c^2 = \pi d_c^2/4,$$

and hence,

$$R_c = d_c/4,$$

where R_c is the blast radius when its strength has decayed to the Chapman-Jouguet value (i.e. $R_s \rightarrow R_c$, $M_s \rightarrow M_{CJ}$, where M_s is the shock Mach number). Using strong-blast theory, one can easily determine the blast energy as a function of the blast radius, and the resultant expression is given by

$$\begin{aligned} E_0 &= 4\pi\gamma p_0 M_{CJ}^2 I (13\lambda/4)^3, \\ &= (2197/16)\pi\rho_0 I D^2 \lambda^3, \end{aligned} \quad (5)$$

which is quite similar to Equations (2b) and (4). Using the cell-size data of Knystautas et al. (1982, 1983), the critical energies have been computed for the common fuel-air mixtures where experimental data are available (J. E. Elsworth, private communication). The results indicate that Equation (5), based on the surface-energy concept, gives the best correspondence. Figure 7 shows a comparison of the predicted results using Equation (5) with those obtained by Elsworth from direct experimental measurements of the critical weight. The critical energy in Figure 7 has been expressed as an equivalent charge weight of tetryl (1 g tetryl is equivalent to 4270 J) for easy comparison with Elsworth's experimental data. The agreement, in general, is reasonably good in view of the simplicity of the model. No experimental data of critical energies have been reported for C_2H_2 -air mixtures to permit a comparison with the present prediction from Equation (5). For most practical situations, Equation (5) can be used to predict critical initiation energies (or charge weights) with quite acceptable accuracy.

DETONABILITY LIMITS

Composition limits refer to the minimum and maximum fuel concentration in which a self-sustained detonation can propagate. From the previous

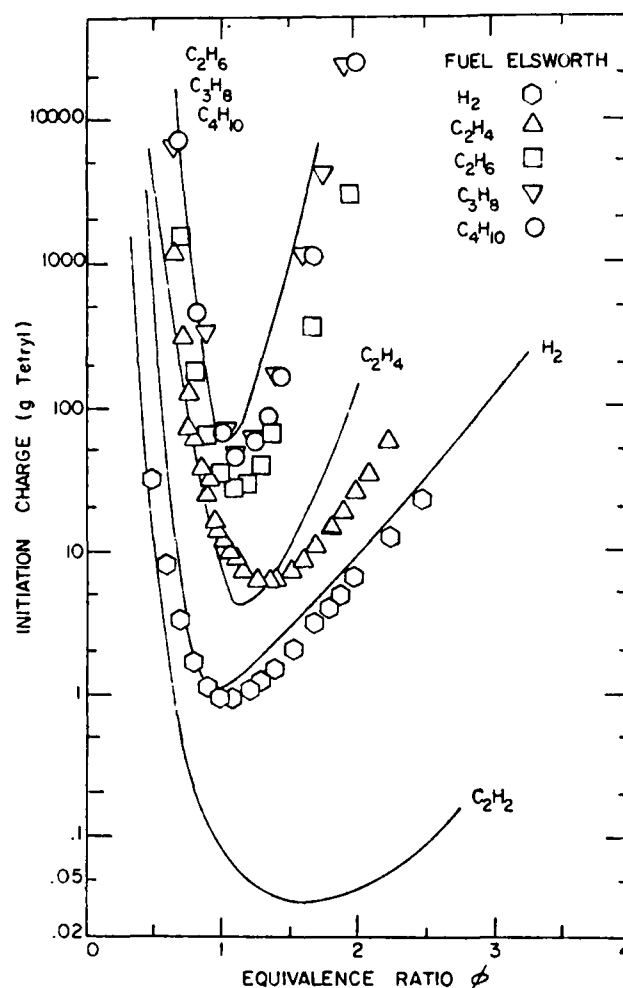


Figure 7 Comparison of critical-energy data for fuel-air mixtures with the surface-energy theory.

discussion, it is clearly meaningless to specify the composition limit without giving the details of the boundary conditions or the environment in which the wave propagates. Thus, for a planar detonation propagating in a rigid tube of circular cross section, the tube diameter must be specified simultaneously with the composition limits. Strictly speaking, the nature of the wall also influences the detonation propagation. Thus, the wall material and its surface roughness must also be specified. In general, different tubes of different diameters, or of different geometrical cross-sectional area, wall roughness, etc., will all have their respective lean and rich limits. Cylindrical and spherical detonations accordingly will also have their particular limiting concentrations. It appears that to be rid of the boundary problem completely, only cylindrical or spherical detonation limits can be considered as "true limits" characteristic of the mixture. We shall return to our discussion of the limits for unconfined detonations later.

Consider the case of "smooth" circular tubes where only the tube diameter need be specified in relation to the limiting composition. There exists the problem of an experimentally derived operational definition of the limits. Since very strong initiators must be used for the near-limit mixtures, the first question to be resolved is how long must the tube be before the influence of the initiator can be considered negligible and the wave is truly self-sustained. Experiments indicate that the effect of the initiator takes an extremely long time to decay (e.g. hundreds of tube diameters). There are strong indications that in confined tubes the single-head spinning mode can be initiated in mixtures outside the limits for a given tube size if a powerful igniter is used. Once initiated, the single-head spinning detonation does not appear to decay at all as it propagates down the tube. It is analogous to an unstable rocket motor resonating at the fundamental transverse mode of instability as it travels down the tube at the Chapman-Jouguet velocity. The experiments of Wolanski et al. (1981) on methane-air detonation are an example of this, since the 5-cm-diameter tube used is much smaller than that required for the propagation of methane-air detonations. Yet, no noticeable attenuation has been observed, and over a wide range of methane concentration, the detonation propagates at the fundamental spinning mode. The existence of such phenomena makes it extremely difficult to establish the limits experimentally. Perhaps there correspond certain limits where, irrespective of the strength of the igniter, a supersonic wave at the fundamental spinning mode propagating at about the Chapman-Jouguet velocity is not possible. However, there exists a range of composition between the first onset of the fundamental spinning mode in a given tube and the composition when it is impossible to initiate a supersonic combustion wave. The question then arises as to whether the supersonic combustion waves in this range are truly self-sustained detonations. That the single-head spinning detonation is the lowest possible stable mode in a given tube (thus the limit) was proposed long ago by Dove & Wagner (1960), among others. However, the recent study by Donato (1982) perhaps sheds some light on this question of what is considered as truly self-sustained single-headed spin. In studying the stability of the near-limit detonations, Donato found that by adding a finite perturbation (such as a few turns of a wire spiral inside the tube) to destroy the spinning wave, the detonation will recover and continue to propagate as a spinning wave only when the composition of the mixture corresponds to the first onset of single-headed spinning detonation in that particular tube. In other words, although the use of a stronger igniter can initiate a single-head spinning detonation wave below the composition when spinning detonation is first observed, the wave is not stable to the finite perturbation by the wire spirals. Once destroyed, the wave will not transit back to the

detonation mode. The more recent studies by Lee et al. (1983) of turbulent-flame accelerations in tubes and transition to detonation confirm the results of Donato. It was found that transition to detonation is only possible in mixtures bounded by the composition limits that correspond to the first onset of single-head spin. In other words, transition to detonation is not possible in mixtures where the composition is poorer than that corresponding to the onset of single-headed structure. For the onset of a single-head spin, the tube circumference corresponds to one complete cell λ . Thus, we have $\pi D = \lambda$, which for a given tube diameter is the critical value of λ . Hence, the corresponding mixture composition can be determined. For example, for stoichiometric H_2 -air mixtures, $\lambda = 1.5$ cm. Thus, the minimum tube diameter that could sustain a stable spinning detonation would be $D \simeq 0.5$ cm. For the alkanes, the minimum diameter would be of the order of $D \simeq 1.7$ cm for stoichiometric composition. It is of interest to note that according to the cell size reported by Moen et al. (1983) for methane (i.e. $\lambda \simeq 33$ cm), a minimum tube diameter for stable detonation propagation in stoichiometric methane-air mixtures would be of the order of at least 10 cm. Thus, the results reported by Wolanski et al. (1981) in a 5-cm tube should correspond to overdriven transient waves only. This is supported by the fact that they observed single-head spinning waves over the whole range of fuel concentrations studied. Based on stability considerations, the composition limit can now at least be defined experimentally.

With the cell size known as a function of composition, the limits for any given circular tube can now be specified by the criterion $\lambda = \pi D$. For two-dimensional planar channels, Vasiliev (1982) recently found that the critical condition for a stable wave corresponds to a channel width w equal to λ . Thus, $w = \lambda$ represents the criterion for two-dimensional rectangular channels of large aspect ratios. What remains to be done is an experimental study of the conditions for stable propagation in tubes of arbitrary geometries. It appears that a unified criterion for limits in tubes can be derived on the basis of the minimum wave-curvature concept discussed earlier.

The question of the limits for unconfined cylindrical and spherical detonations is much more difficult. Here there are no boundaries to be considered, and the answer must lie within the detailed processes of the cellular structure. In particular, the limits must be determined from the conditions whereby the cells can multiply in an unconfined wave. Stable propagation requires that the averaged cell size be constant for a given mixture. Hence, the detonation cells must multiply in a diverging wave front in accordance with the rate at which the surface area increases. The mechanism for the multiplication of the cells is in the transient processes of

double Mach reflections (in particular, how the second Mach stem is formed and develops to form a new cell). Even in nonreacting media, the problem of describing various types of triple-shock interactions has not been fully resolved. For the cellular-detonation case, further complexities are introduced by the addition of flame fronts into the already complex shock pattern. The interaction process is also highly transient, and similar types of approximations may not apply. Apart from the fundamental limit mechanism of cell reproduction for unconfined detonations, it is also difficult to formulate the necessary operational definition for the experimental measurements of limits for diverging waves. It appears that the current method of arbitrarily specifying an upper initiation energy above which the mixture is rendered nondetonable is reasonable, at least from a practical point of view. As such, the cell size also enters through the initiation energy. Specifying an upper value for the initiation energy is then equivalent to specifying a maximum value of the cell size beyond which the mixture is too insensitive to be detonable. Although there are other means of specifying reasonable limits for unconfined detonation from the practical point of view, such as the use of the size of the detonation kernel as the minimum detonable volume, the limit problem remains fundamentally linked to the nature of the cellular structure.

CONCLUSIONS

The cell size has been demonstrated to be the most fundamental parameter characterizing the dynamic detonation properties. It has also been demonstrated that a knowledge of the detonation cell size permits the dynamic parameters to be determined reasonably well. However, there is much room for refining the existing relationships that link λ to these dynamic parameters. The 13λ correlation between the cell size and the critical tube diameter d_c is presently a purely empirical relationship. Its validity is based on experiments on the common hydrocarbon fuels. Although the ranges of initial conditions and mixture compositions studied are fairly wide, all the fuels studied have similar kinetics of hydrocarbon oxidation. Thus it seems worthwhile to test the $d_c = 13\lambda$ correlation in other detonable systems, such as H_2-Cl_2 or CS_2-O_2 mixtures, where other fuel molecules are involved. Also, if the 13λ correlation is related to the concept of an effective or hydrodynamic thickness of the detonation wave, then the use of various inert diluents to modify the sound speed in the product gases should influence the equilibrations of the transverse wave and, hence, the hydrodynamic thickness. This may have an effect on the correlation $d_c = 13\lambda$. Strehlow (1968) has already demonstrated that the use of heavy argon dilution has significant influence on the "regularity" of

the cellular structure of $\text{H}_2\text{-O}_2$ detonations. Further study of the transmission from different geometries, as well as the transmission from rigid to flexible wall tubes, could provide additional insight into the physics behind the $d_c = 13\lambda$ correlation. A correct theoretical model to explain this simple correlation will pave the way to the development of quantitative theories for the other dynamic parameters.

Regarding the prediction of λ from the given kinetics and physical properties of the mixture, it is the opinion of the author that "brute force" numerical computations will not lead to the solution of the problem. The feasibility of numerical simulations and the propagation of two-dimensional cellular detonations have already been demonstrated. The extension to the simulation of three-dimensional cellular detonations is, in principle, possible. It is, however, not clear what additional physics can be learned from these complex computations. Certainly, the use of these codes to compute the cell-size data from the basic kinetics of the system is not feasible from the economics standpoint. It would be much simpler to obtain cell-size data experimentally. There is, however, an urgent need to improve both the technique and accuracy for cell-size measurements and to eliminate the "experience factor" in the identification of the dominant cell size in an irregular pattern. If a relationship between the cell size and another more easily measured dynamic parameter can be firmly established, then perhaps the cell size can be determined indirectly from the measurement of this parameter.

Thus, it seems that if λ can be treated as a fundamental parameter characterizing the detonation mode of combustion, then we are well on the way to developing predictive theories for the important dynamic parameters of practical interest. The cell size λ , like the fundamental kinetic-rate constants of the elementary reactions, can be considered as a fundamental property of the mixture. Adopting such a point of view, the theory of gaseous detonation can be said to be very close to completion after a century since the initial discoveries.

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Thus, it seems that if λ can be treated as a fundamental parameter characterizing the detonation mode of combustion, then we are well on the way to developing predictive theories for the important dynamic parameters of practical interest. The cell size λ , like the fundamental kinetic-rate constants of the elementary reactions, can be considered as a fundamental property of the mixture. Adopting such a point of view, the theory of gaseous detonation can be said to be very close to completion after a century since the initial discoveries.

APPENDIX VI

High Speed Turbulent Deflagrations and Transition to Detonation in H_2 -Air Mixtures

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An experimental investigation on flame acceleration and transition to detonation in H_2 -air mixtures has been carried out in a tube which had a 5 cm cross-sectional diameter and was 11 m long. Obstacles in the form of a spiral coil (6 mm diameter tubing, pitch 5 cm, blockage ratio $BR = 0.44$) and repeated orifice plates spaced 5 cm apart with blockage ratios of $BR = 0.44$ and 0.6 were used. The obstacle section was 3 m long. The compositional range of H_2 in air extended from 10 to 45%, the initial pressure of the experiment was 1 atm, and the mixture was at room temperature. The results indicate that steady-state flame (or detonation) speeds are attained over a flame travel of 10-40 tube diameters. For $H_2 \leq 13\%$ maximum flame speeds are subsonic, typically below 200 m/s. A sharp transition occurs at about 13% H_2 when the flame speed reaches supersonic values. A second transition to the so-called quasi-detonation regime occurs near the stoichiometric composition when the flame speed reaches a critical value of the order of 800 m/s. The maximum value of the averaged pressure is found to be between the normal C-J detonation pressure and the constant volume explosion value. Of particular interest is the observation that at a critical composition of about 17% H_2 transition to normal C-J detonation occurs when the flame exits into the smooth obstacle-free portion of the tube. For compositions below 17% H_2 , the high speed turbulent deflagration is observed to decay in this portion of the tube. The detonation cell size for 17% H_2 is about 150 mm and corresponds closely to the value of πD that has been proposed to designate the onset of single-head spinning detonation, in this case for the 5 cm diameter tube used. This supports the limit criterion, namely, that for confined detonations in tubes, the onset of single-head spin gives the limiting composition for stable propagation of a detonation wave.

1. INTRODUCTION

The study of flame propagation through an array of orifice plates arranged in periodic sequence along the length of a flame tube was pioneered by Chapman and Wheeler [1] more than 50 years ago. In this early work, the rapid acceleration of the flame as a result of the action of such restrictive obstructions to velocities in excess of 400 m/s in premixed methane-air mixtures was observed. In the midseventies Wagner [2] extended the preliminary nature of Wheeler's early study to a more exhaustive parametric investigation in

which the effects of blockage ratio and orifice spacing were considered. Wagner's work indicates that under optimal conditions, flame velocities in excess of 700 m/s for stoichiometric CH_4 -air mixtures can be achieved just beyond a short array of 21 equispaced obstacles in a 4 cm diameter tube. Subsequently, research on turbulent flame acceleration by repeated obstacles in one-dimensional channels and in cylindrical diverging configurations has been carried out at McGill University [3, 4] and extended to fairly large scale in collaboration with Eckhoff at the Chr. Michelson Institute [5] ($2\frac{1}{2}$ m diam \times 10 m long tube) and with Wagner at the Physical Chemistry Institute [6] ($2\frac{1}{2}$ m \times $2\frac{1}{2}$ m channel). However, in all these previous studies, the flame tubes

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were too short on a relative scale (L/D small) to permit steady-state conditions to be achieved at the end of the flame travel. Hence, the maximum turbulent flame speed for a given mixture and obstacle configuration was not reached, and important information on whether transition to detonation is possible could not be deduced from these experiments. From a fundamental point of view, it is of great interest to explore the mechanisms involved in curbing the powerful flame acceleration mechanism to yield eventually an optimal turbulent burning rate. It is also of great interest to examine the propagation mechanism of very high speed deflagrations which may lead to the understanding of the critical conditions underlying the transition process to the detonation regime. From a practical point of view, information on maximum burning rates which would permit the estimation of pressure development in explosions, run-up distances, and critical conditions for transition to detonation is of importance in the context of probabilistic risk assessment. The present paper reports some recent experimental results on flame acceleration in long tubes ($L/D > 200$) in the presence of repeated obstacles. Hydrogen-air mixtures have been used because they are sufficiently sensitive so that run-up distances to the steady state are short and transition to detonation can be studied over a wider range of H_2 concentrations than less sensitive hydrocarbon-air mixtures (e.g., CH_4). The

interest in hydrogen deflagrations has additional impetus in view of the current concern of explosion hazards in light water nuclear reactors due to the possibility of large scale hydrogen release following loss of coolant and core meltdown-type accidents, exemplified by the Three Mile Island incident in 1979.

2. EXPERIMENTAL DETAILS

A steel combustion tube 5 cm in diameter and 11 m long ($L/D = 220$) was used in the present experiments. The tube was equipped with three types of turbulence inducing obstacles: (i) a spiral coil (Shchelkin spiral) made from 6 mm diameter tubing with a pitch of 5 cm and effective blockage ratio $BR = 1 - (d/D)^2 = 0.44$; (ii) circular orifice plates with $d = 37.5$ mm, $BR = 0.44$, and 5 cm plate separation; (iii) circular orifice plates with $d = 31.6$ mm, $BR = 0.6$, and also 5 cm plate separation (Fig. 1). The length of each obstacle section was 3 m and the obstacles were located to start at the ignition end. Thus there was an 8 m section of smooth obstacle-free tube after the flame left the obstacle region. Ionization probes spaced at 50 cm intervals along the tube lengths were used for flame speed measurement. The probe tips were extended to the axis of the tube, and thus the flame speeds reported correspond to those along the tube axis. The ionization gauges could be replaced by piezoelectric pres-

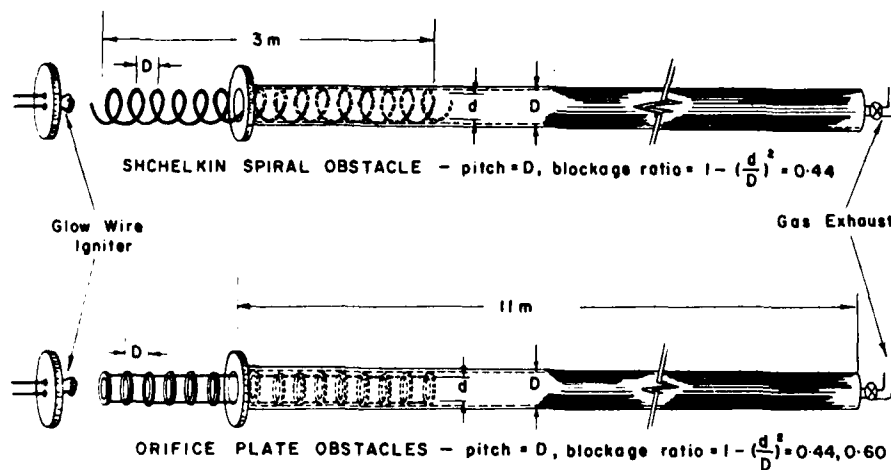


Fig. 1. Schematic diagram of the flame tube and obstacle arrays.

sure transducers (PCB 113A24) for recording the pressure-time development. The signals were recorded using a Biomation digital waveform recorder (Model 805) to permit the control of signal amplitude and time scales for data analysis. For most experiments, ignition of the mixture was achieved by a glow plug igniter; in weaker mixtures, a more powerful electric spark discharge was used to improve the reliability of initiation. The explosive mixture was prepared in a continuous flow system, with the desired composition achieved by regulating the respective flow rates of hydrogen and air. Standard calibrated rotameters (Gilmont G-2155) were used to monitor the flow rates. To ensure a homogeneous mixture, the gases were flowed through a mixing chamber equipped with baffle plates prior to entering the combustion tube. The tube was

first evacuated, and then no less than 5 tube volumes of the explosive mixture were flushed through the tube before each experiment. The initial pressure of the experiments was atmospheric, and the temperature was the ambient value. The range of H_2 concentrations in the present study was $10\% \lesssim H_2 \lesssim 45\%$.

3. RESULTS AND DISCUSSION

The variation of the flame speed (relative to laboratory coordinates) with distance along the combustion tube for the case of the spiral obstacle (pitch equals diameter; blockage ratio $BR = 0.44$) is shown in Fig. 2 for various H_2 concentrations. Although the total tube length was 11 m, with the first 3 m constituting the obstacle field, only the first 8 m were equipped for measurement.

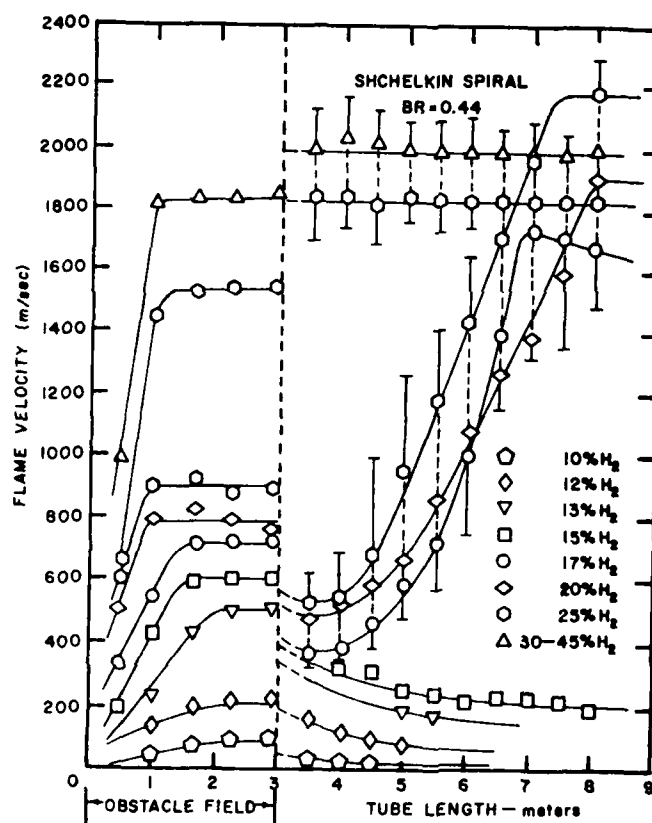


Fig. 2. Variation of flame velocity along the flame tube when a Shchelkin spiral obstacle array ($BR = 0.44$) is used in the first 3 m of the tube.

Within the obstacle field, for $H_2 \leq 17\%$, the deflagration was observed to accelerate to a steady state after about 1.5–2 m (or 30–40 tube diameters) of travel. However, for higher concentrations of H_2 , the steady state was achieved in less than 1 m for the present case of the spiral obstacle. It is of particular interest to note that there appears to be a sudden increase in the steady velocity from about 200 m/s for 12% H_2 to almost 500 m/s at 13% H_2 . This sharp increase in the flame speed occurring at around 13% H_2 is common to all three obstacle configurations investigated. An explanation that suggests itself that can possibly account for this rather sudden transition is that there is a changeover in the dominant kinetic step of the chemical reaction. As pointed out by Atkinson et al. [7], among others [8, 9], the reaction $H + O_2 \rightarrow OH + O$ is in competition with the three-body reaction $H + O_2 + M \rightarrow HO_2 + M$, with the latter dominating at temperatures below about 1350°K. At about 13% H_2 , the flame temperature is of the order of 1300°K. Thus, a changeover to the more rapid OH branching reaction may take place and result in a rapid increase in the maximum turbulent flame speed observed at 13% H_2 . A second sharp transition is also noted at about 25% H_2 , when the flame speed jumps from about 800 to 1500 m/s. In fact, 25% H_2 seems to be the critical composition in which the flame speed fluctuates between two regimes randomly from experiment to experiment. The high velocity regime corresponds to the regular detonation mode of combustion, but the pressure losses suffered by the detonation in propagating through the obstacles account for the large velocity deficit observed (i.e., 1500 m/s, as compared with the normal C-J value of 1861 m/s for 25% H_2). These low velocity detonations resulting from severe pressure losses inflicted by the obstacles are henceforth referred to as quasi-detonations. Their propagation mechanism is strongly influenced by shock reflections [10] and violent turbulent mixing caused by the obstacles. This differs from the usual transverse wave collision mechanism responsible for the propagation of the normal cellular detonations with velocities quite close to the theoretical C-J values.

The sudden transition to the quasi-detonation regime suggests that some critical value for the flame speed needs to be reached before the conditions for the onset of detonation can occur. For the case of the spiral obstacle, the steady flame speed increases with H_2 concentration, and at 25% H_2 a value of about 800 m/s is attained. Such a flame speed drives a shock wave of about $M_s \cong 2.7$, which when reflected from the obstacles yields temperatures of the order of 1200°K. This corresponds closely to the auto-ignition temperatures for shock heated mixtures. These results suggest that the necessary condition for the sudden transition is associated with a requirement for some minimum critical value of the flame speed. This is compatible with previous studies of transition from deflagration to detonation in tubes which indicate certain critical threshold values for the flame speed prior to the onset of detonation. The various modes of transition to detonation revealed in the classical paper of Urtiew and Oppenheim [11] all indicate the necessity for a certain minimum flame speed for the onset of detonation.

To verify the above suggestion that a minimum flame speed is required for transition, experiments were carried out by propagating a normal C-J detonation initiated in the obstacle-free section back into the obstacle region (designated as backward propagation). A hysteresis effect is noted (e.g., at 20% H_2 shown in Fig. 3) in that when a fully established C-J detonation is propagated back from the obstacle-free into the obstacle-filled section, it settles down to a velocity of about 1200 m/s, as compared with the maximum steady-state turbulent flame speed of about 800 m/s that is attained via the continuous flame acceleration process (designated as forward propagation). Under the conditions existing in the present experiment, transition to detonation via continuous flame acceleration does not occur, possibly due to the fact that the critical value of flame speed required for transition is not attained. However, when started out as a fully developed C-J detonation prior to entering the obstacle region, the velocity is seen to decay to the lower quasi-detonation value corresponding to the obstacle configuration and mixture composition. For mix-

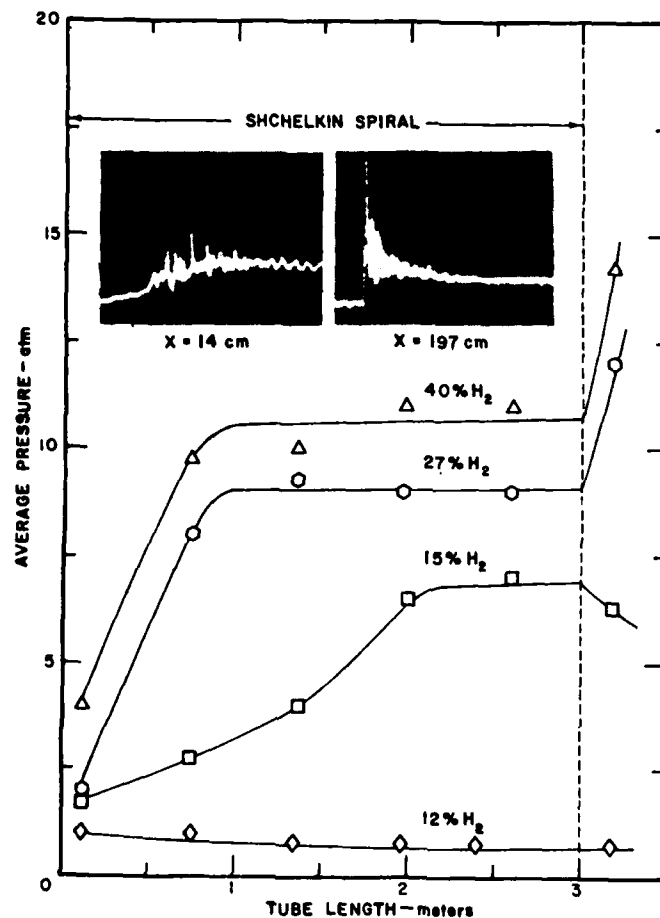


Fig. 3. Variation of pressure along the flame tube and within the Shchelkin spiral obstacle array ($BR = 0.44$).

ture compositions where transition from flame to quasi-detonation can occur in the obstacle region (i.e., the critical value of the flame speed is attained), no hysteresis effect is observed. The quasi-detonation velocity obtained is identical for both cases, whether the starting point is a normal C-J detonation or a flame undergoing transition to a quasi-detonation later. This occurs for mixtures in the range $25\% < H_2 < 45\%$ for the (Shchelkin) spiral obstacle as shown in Fig. 3.

Observations downstream of the obstacle section when the flame or quasi-detonation propagates into the obstacle-free portion of the tube also yield some interesting results. From Fig. 2 it can be noted that below 17% H_2 , the flame

speed decays rapidly downstream of the obstacle section. This is due to the absence of obstacles to randomize the mean flow to produce the level of turbulence intensity required to sustain the high burning rate. Below about 13% H_2 , the flame speed decays drastically to a few tens of meters per second within about 20–40 tube diameters. For $H_2 \geq 13\%$, the flame speed first suffers a sharp initial drop, then decays slowly to about a couple of hundred meters per second. At compositions for $H_2 \geq 17\%$ there clearly is a dramatic change in the nature of the flame decay phenomenon. After the sharp initial drop the flame is seen to reaccelerate and undergo transition to a normal C-J detonation after a few meters of travel. The

data indicate that there is considerable scatter of results in this region typically symptomatic of the abruptly sporadic nature of the transition process. Below 17% H_2 , the flame simply decays monotonically to low velocity with increasing distance. Thus, 17% H_2 appears to be the critical concentration that gives rise to transition to normal detonation in the obstacle-free portion of the 5 cm diameter tube. However, inside the obstacle filled section with the particular spiral obstacle configuration used, 25% H_2 appears to be the critical concentration for which transition to a quasi-detonation wave is seen to occur within the obstacle section. In examining the detonation cell size for H_2 -air mixtures [12], it is found that for 17% H_2 , the cell size is about 15.0 cm. This corresponds very closely to the value of πD for the detonation cell size for mixtures at the onset of the single-head spinning mode in a circular tube of diameter D . This confirms the criterion proposed for detonation limits in circular tubes that the critical composition should be based on the onset of single-head spinning detonations [13]–[15]. This limit criterion was proposed on the basis of experimental observations [15] that only mixtures whose cell sizes are less than that corresponding to the single-head spin in the given tube permit the propagation of a stable detonation. A stable detonation in this case means one which, having been destroyed by a finite perturbation, such as that induced by blockage in the tube, say, via an orifice plate, will rapidly undergo transition back to a normal detonation. In the context of the above definition of detonability limits, it is of interest to refer to the work of Wolanski et al. [16] on methane-air detonations in a 5 cm tube which yielded single-head spin waves for all CH_4 concentrations. Clearly, their experimental conditions are beyond the composition limit for the onset of single-head spin, and the wave is not stable to finite perturbations. Once the detonation wave is destroyed, it will never recover to propagate in the detonation mode again. Such waves are formed in the first place due to the use of very powerful igniters. The present result is therefore significant in that it demonstrates that if the mixture composition is within the detonability limits for the given tube,

then it is possible for transition from deflagration to detonation to take place. It is evident that when and if transition occurs depends on the effectiveness of the flame acceleration mechanisms available to bring the flame speed up to meet certain critical requirements.

The corresponding pressure variation with distance from the igniter for the Shchelkin spiral obstacle array is shown in Fig. 4 together with two typical pressure traces: one corresponding to a pressure measurement close to the igniter at the very beginning of the obstacle array, the other roughly two-thirds of the way along the array. One can immediately note that the pressure front steepens with increasing distance along the obstacles as shown on the oscilloscope traces; this is qualitatively similar to the trend observed by Wagner [2] for methane-air flames. However, in the present case of the hydrogen-air flames, the pressure-time records exhibit strong oscillations over and above the baseline mean pressure changes. This is due to the interaction of the flow with the periodically spaced obstacles and is compatible with the wave pattern shown in the streak records reported by Wagner [2] for a flame propagating in a regularly spaced obstacle array. The pressure plotted in Fig. 4 refers to the maximum mean value. The time to reach this maximum mean value of the average pressure ranges from 10 ms or more for very lean mixtures (e.g., $H_2 \cong 12\%$) to a few microseconds in the quasi-detonation regime ($H_2 \geq 25\%$). For very lean mixtures, the maximum pressure obtained is the same for all the transducer locations. This is due to the fact that the flame speed is less than the sound speed (~ 350 m/s), which is a measure of the rate at which the local pressure inhomogeneities generated by the flame are dispersed and equalized. For $H_2 \geq 13\%$, where the flame speed becomes supersonic, local pressure buildup is dispersed at speeds of the order of the flame speed. Thus, the pressure front closely coincides with the flame front and the approach to steady state for both the flame speed and the maximum pressure become identical, as indicated in Fig. 4. Beyond the obstacle region, the pressure will either rise further or fall, depending on whether the flame or quasi-

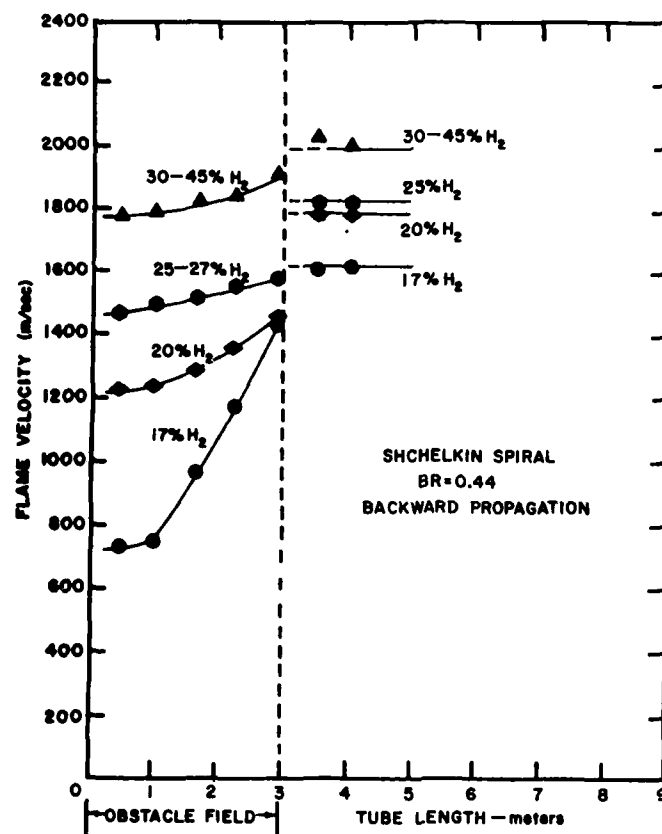


Fig. 4. Variation of flame velocity along the flame tube for the Shchelkin spiral obstacle array, with a blockage ratio, $BR = 0.44$, for backward propagation.

detonation undergoes transition to a normal C-J detonation or decays continuously to a relatively low speed deflagration compatible with the obstacle-free portion of the tube.

A comparison of the flame developments for the three obstacle configurations of spiral ($BR = 0.44$), orifice plate ($BR = 0.44$), and orifice plate ($BR = 0.6$) can be seen from Figs. 2, 5, and 6. In terms of flame acceleration rates, orifice plates are more efficient than the spiral, for the same blockage ratio, even though this may not be a meaningful comparison, since the spiral has a different geometry. However, for the same orifice plate configuration, a higher blockage ratio gives rise to steady-state conditions much earlier. The steady-state flame speeds and/or quasi-detonation velocities are weakly dependent on the obstacle

configuration and blockage ratio. However, the first sudden jump in the flame speed occurring at about 13% H_2 is common to all three obstacle configurations, indicating that the kinetic explanation is a plausible one. Furthermore, the reacceleration of the flame to detonation after leaving the obstacle section at the critical compositions above about 17% H_2 for the three cases also confirms the single-head spin limit criterion discussed earlier. A more convenient comparison of the steady-state flame speeds for various H_2 concentrations for the three obstacle configurations is given in Fig. 7. Here, the first velocity jump in the flame speed at about 13% H_2 for all the three cases as well as subsequent dependence of the flame speed on blockage ratio for different H_2 concentrations can be clearly observed. The

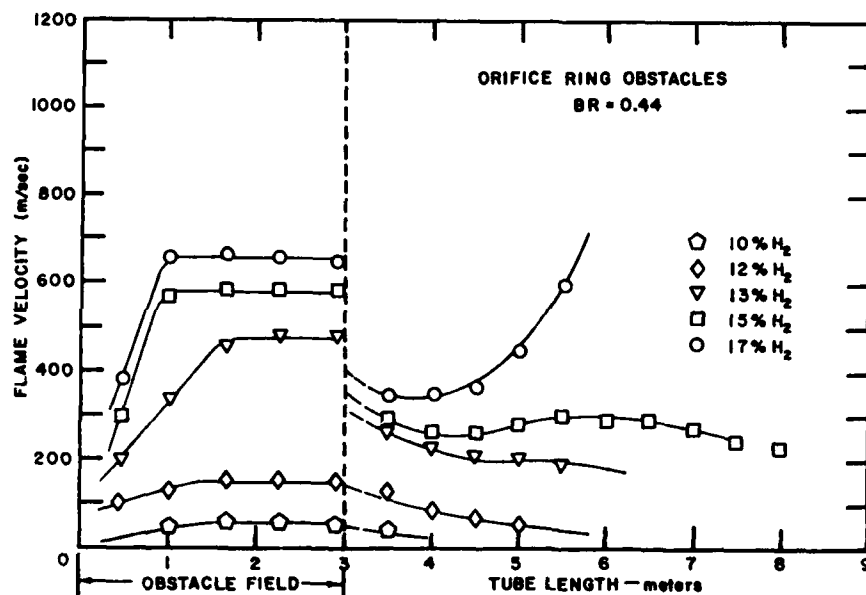


Fig. 5. Variation of flame velocity along the flame tube for the orifice ring obstacle array with a blockage ratio $BR = 0.44$.

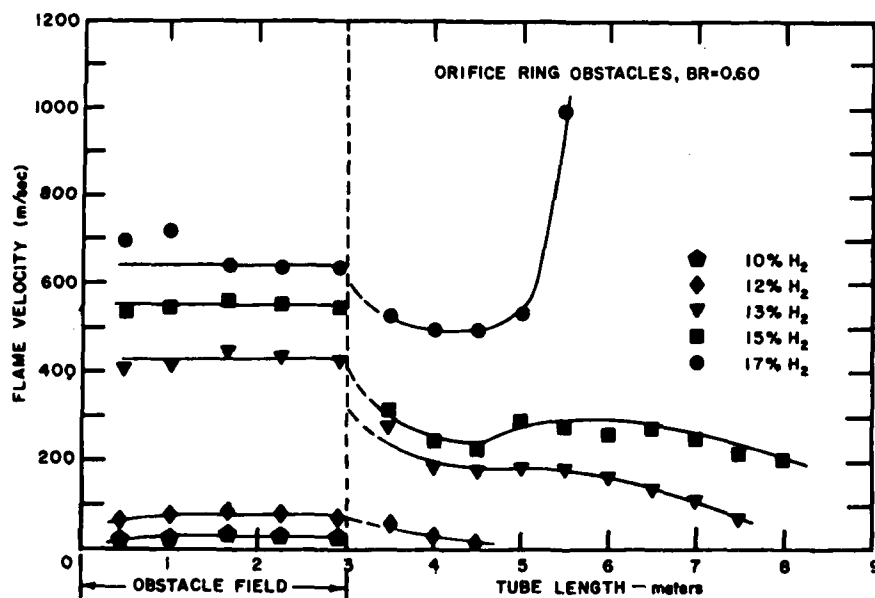


Fig. 6. Variation of flame velocity along the flame tube for the orifice ring obstacle array with a blockage ratio $BR = 0.60$.

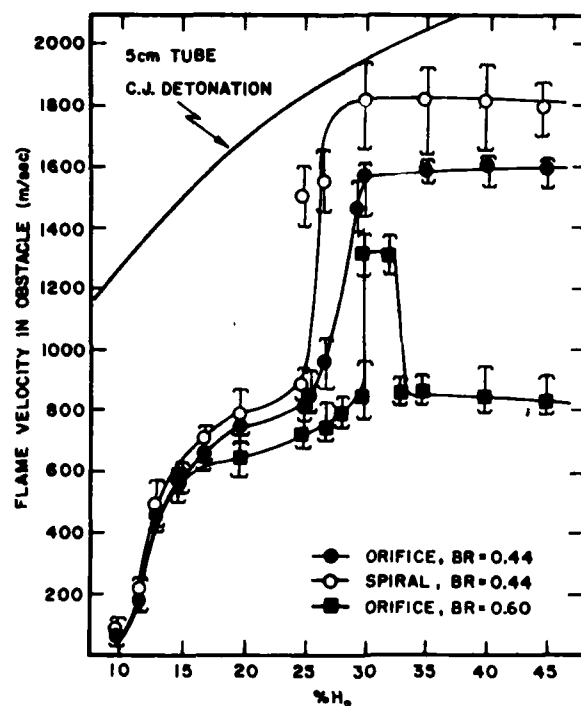


Fig. 7. Variation of the maximum steady-state flame velocity with composition of the H_2 -air mixture for the three obstacle array configurations studied.

second jump, corresponding to the transition to the quasi-detonation regime, is also common to all cases. It is of particular interest to note that the flame speed just prior to the transition is about 860 m/s for all three cases. This confirms the notion that perhaps a critical value of the deflagration speed is required before transition from deflagration to detonation can take place. For different obstacle configurations and blockage ratios, this critical value is attained at different H_2 concentrations. For example, with the large blockage ratio of 0.6, the pressure losses are sufficiently severe to limit the steady-state flame speed to a value below the critical value until almost the stoichiometric composition is reached. However, for the spiral configuration, where the pressure losses are much less severe, the flame accelerates more readily and the critical value required for transition can be reached at a lower H_2 concentration of about 25% H_2 . It is possible that for very dense obstacle arrays with sufficiently

high pressure losses, transition to detonation may be suppressed entirely while propagating in the obstacle field, since the maximum flame speed attained may be below the required level. Presumably, the critical value of the flame speed must also depend on the H_2 concentration as well as other parameters. In the present study, about 800 m/s appears to be the minimum critical value of flame speed required for transition to detonation. For H_2 -air mixtures such a value of flame speed corresponds roughly to H_2 concentrations around the stoichiometric composition.

For the higher blockage ratio of 0.6, transition to detonation again is suppressed for rich mixtures with $H_2 \gtrsim 33\%$. Thus, for this particular obstacle configuration, transition is observed only in the narrow range of $30\% \lesssim H_2 \lesssim 33\%$. For lower blockage ratios (i.e., $BR = 0.44$), transition is observed for rich mixtures up to 45% H_2 . The implication here is that eventually, for even richer

mixtures (i.e., $H_2 \gtrsim 45\%$), transition again would not be possible for the lower blockage ratio configurations.

It is of interest to note that in the range $25\% \lesssim H_2 \lesssim 45\%$ for the case of $BR = 0.44$, the quasi-detonation velocity remains more or less constant with composition, even though the normal C-J detonation velocity shows a continuous rise with increasing H_2 concentration. This indicates the strong influence of pressure losses generated by the obstacles on the propagation of these quasi-detonations. Increasing the detonation velocity with H_2 concentration increases the momentum losses, thus maintaining a more or less constant value of the quasi-detonation velocity itself.

However, for different obstacle configurations and blockage ratios, the value of the quasi-detonation velocity varies. For example, for the spiral the quasi-detonation velocity is around 1800 m/s in the range $25\% \leq H_2 \leq 45\%$, while for the orifice plates with the same blockage ratio of 0.44, the quasi-detonation velocity is about 1600 m/s. This indicates that the pressure losses associated with the orifice plates are higher. For the orifice plates with $BR = 0.6$, the quasi-detonation velocity is only about 1300 m/s.

The maximum mean pressure measured at 197 cm from the ignition end is plotted in Fig. 8 against H_2 concentration for all three obstacle configurations. From previous figures, we note

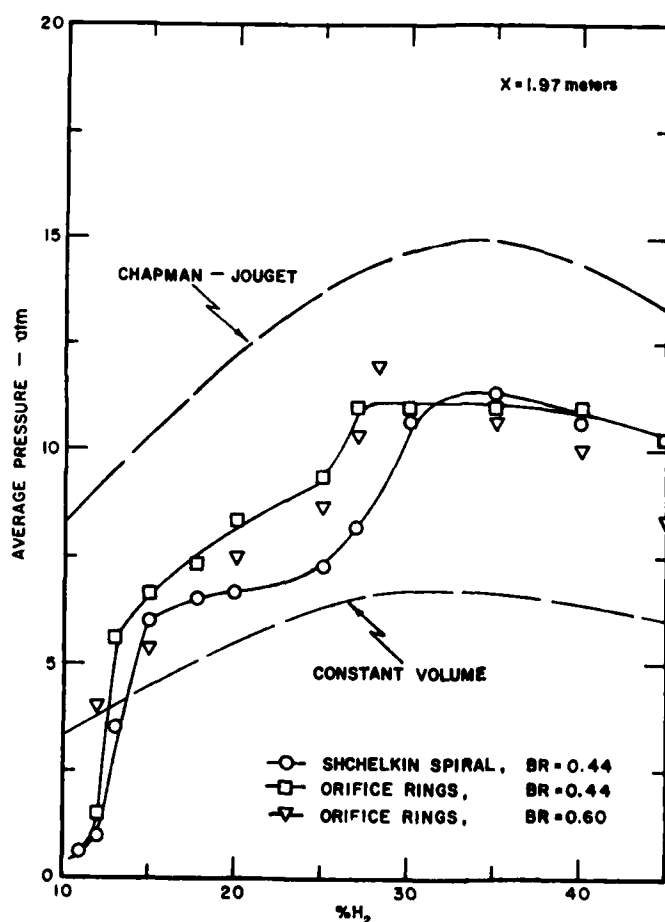


Fig. 8. Variation of the maximum mean pressure with composition of the H_2 -air mixture for the three obstacle array configurations studied.

that steady-state conditions (either turbulent deflagrations or quasi-detonations) are achieved at this location. In Fig. 8, we again note the rapid increase in pressure for all obstacle configurations at about 13% H₂. Prior to transition to the quasi-detonation regime, the maximum mean pressure is found to be slightly above the theoretical constant volume explosion pressure. Dependence of the pressure on obstacle configuration is weak throughout. In the range of compositions where transition from high speed deflagration to quasi-detonation (i.e., $25\% \lesssim \text{H}_2 \lesssim 30\%$) occurs, the pressure rises only slightly to values about midway between the equilibrium C-J detonation pressure and the constant volume explosion pressure. Unlike the flame speed, which shows a more distinct jump when transition to quasi-detonation occurs, the pressure shows only a slight increase. In the quasi-detonation regime, the spiral and orifice plate obstacles with the same blockage ratio give almost identical pressures even though the detonation velocity shows a distinct difference of about 200 m/s. For the higher blockage ratio plates of BR = 0.6, the quasi-detonation pressure is also practically identical to the other configurations ($\text{H}_2 \lesssim 34\%$). For richer mixtures (i.e., $\text{H}_2 \gtrsim 34\%$), where Fig. 7 indicates no transition to quasi-detonation for BR = 0.6, the pressure for the high speed deflagration regime decreases only slightly from the corresponding quasi-detonation values. In fact, it may be concluded that as far as the pressure is concerned, little difference is observed between the high speed deflagration and the quasi-detonation regimes when the blockage ratio or pressure losses are sufficiently high. The distinction becomes more prominent when the pressure losses are relatively small (e.g., the spiral) and the two regimes can be separated more clearly when transition occurs.

4. CONCLUSIONS

The results of the present investigation indicate that both chemistry and gas dynamics play important roles in the control of freely propagating turbulent deflagrations. For mixtures near the limits where the reaction rates are low, it appears that turbulent quenching is responsible for the

upper bound of the burning rate and hence of the deflagration speed. In other words, the self-acceleration process will bring the flame up to velocities where the turbulent mixing rate becomes of the same order of magnitude as the reaction rate. Further increase in turbulence intensity will result in quenching rather than an augmentation of the combustion processes. Equality between some characteristic eddy life time and the global chemical reaction time would be an appropriate criterion for this regime.

For sufficiently short reaction times, quenching will not set in until the displacement flow generated by the flame starts to choke gas dynamically from the pressure losses due to the obstacles. Thus, frictional choking now poses the upper limit rather than the chemical reaction rate, and the maximum deflagration speed will depend on the losses as well as the power output of the combustion processes that is responsible for the generation of the displacement flow itself. Choking or sonic flow conditions present the criterion for this gas dynamic regime. For the present case of H₂-air mixtures, the sudden increase in reaction rate at around 13% H₂ leads to a distinct transition from the turbulent quenching to the gas dynamic choking regimes. In other mixtures where such a distinct change in the chemical reaction rates with composition is absent, it would be difficult to discern the two regimes. The onset of sonic flow conditions would perhaps signify that choking is present and that the gas dynamic mechanism begins to play a more prominent role.

A second distinct regime, the so-called quasi-detonation regime, is also observed. The onset of the quasi-detonation regime is marked by a sudden jump in the velocity of the turbulent deflagration, although the averaged maximum pressure does not demonstrate such a distinct difference between the two regimes. Depending on the pressure loss, the quasi-detonation velocities can be as low as 30% of the normal C-J values, but the maximum averaged pressure for high speed deflagrations and quasi-detonations is about the same, having values typically midway between the constant volume explosion pressure and the normal C-J detonation pressure. The pressure-time histories for high speed turbulent

deflagrations and quasi-detonations are quite similar, and only the velocity jump marks the transition between the two regimes. Transition to quasi-detonations appears to require a minimum flame speed of the order of 800 m/s. In other words, for a given obstacle configuration, the mixture composition in which transition occurs appears to be governed by the attainment of this minimum flame speed of about 800 m/s.

Regarding acceleration to steady state, it is found that higher pressure losses (i.e., larger blockage ratio) give more rapid rate of flame acceleration. This is compatible with the notion that pressure losses are associated with the randomization of the mean flow into turbulence, which in turn augments the combustion rates. For very lean mixtures (e.g., $H_2 \cong 10\%$), the steady state is achieved very rapidly, suggesting that turbulent quenching is the dominant mechanism and sets in immediately, since the slow reaction rate does not require high turbulent intensities. For more rapidly burning mixtures, the flame acceleration to steady state takes correspondingly longer, since it takes time to build up a sufficiently high gas dynamic flow velocity to produce the required turbulence intensity for the quenching mechanism to be effective. For sufficiently high velocity, frictional choking may set in and present another mechanism that limits the rate of buildup of the mean flow velocity itself. For H_2 -air mixtures, the very rapid increase in the kinetic reaction rates at around 13% via the OH branching step results in a very rapid increase in the turbulent flame speed to sonic velocity, and choking then becomes the dominant mechanism. In other mixtures with a different kinetic mechanism, there may not be a sudden transition, and no sharp jump in velocity would be observed.

Of particular interest are the phenomena downstream of the obstacle section. There appears to be a critical H_2 concentration of about 17% H_2 above which the deflagration upon leaving the obstacle filled section will abruptly undergo transition to a normal C-J detonation. Below this critical concentration, the deflagration decays, since there are no more obstacles to support the

required turbulence level for high speed combustion. Corresponding to the critical composition of 17% H_2 , the detonation cell size is about 156 mm and agrees closely with the value of πD for the onset of single-head spinning detonation in the 5 cm tube used. This is in accord with the criterion that the onset of single-headed spin detonations represents the limiting condition for stable detonation propagation in a given tube. The present results go further in demonstrating that transition is only possible in a given tube if the mixture composition is such that the cell size is less than the πD value for single-head spin. This suggests that establishing detonability limits could be approached by carrying out experiments exploring the possibility for transition itself.

The present results, which demonstrate the high deflagration speed achieved when $H_2 \gtrsim 13\%$, suggest that the lean limit for H_2 -air detonation could be in the neighborhood of 13% H_2 , as suggested by Atkinson et al. [7]. In fact, our recent experiments [17] in a 30 cm diam tube demonstrate that 13.8% H_2 can be detonated with a corresponding cell size of about 60 cm. For 13% H_2 , with an estimated cell size of the order of 80 cm, the minimum tube diameter for transition or stable single-head detonation would be of the order of about 27 cm.

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value of the averaged pressure is found to be between the normal C-J detonation pressure and the constant volume explosion value. Of particular interest is the observation that at a critical composition of about 17% H_2 transition to normal C-J detonation occurs when the flame exits into the smooth obstacle-free portion of the tube. For compositions below 17% H_2 , the high speed turbulent deflagration is observed to decay in this portion of the tube. The detonation cell size for 17% H_2 is about 150 mm and corresponds closely to the value of wD that has been proposed to designate the onset of single-head spinning detonation, in this case for the 5 cm diameter tube used. This supports the limit criterion, namely, that for confined detonations in tubes, the onset of single-head spin gives the limiting composition for stable propagation of a detonation wave.

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